Nonlinear Spectroscopy of Atoms in Magneto-Optical Trap

by

Tomasz M. Brzozowski

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prof. dr hab. Wojciech Gawlik, Supervisor
prof. dr hab. Tomasz Dohnalik, Referee
dr hab. Marek Tripchenbach, Referee

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Abstract

This thesis presents the high-resolution spectroscopic measurements of the $^{85}$Rb atoms confined in a magneto-optical trap (MOT). The absorption and four-wave mixing spectra obtained in the present experiment are a rich source of information on the light-atom interaction in the working MOT, i.e. when all trapping fields remain switched on during the phase of measurement. The observed ultra-narrow resonances in the registered spectra are due to the motion of atoms, hence, the presented spectroscopic methods can be used to diagnose the properties of the cold sample connected with atomic external degrees of freedom. The expansion of the theory of the generic pump-probe spectroscopy configurations by inclusion of realistic MOT conditions leads to the good agreement with experimental data.
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Introduction

Spectroscopy

Spectroscopy, or more precisely - the aim of spectroscopic research - could be defined as “learning physical properties of the matter using light and studying the light-matter interaction”. Even in our everyday life, when we look at a particular object we perform a sort of spectroscopic measurement: the reflected light which enters our pupil gives us a wealth of information about the object. While real spectroscopic techniques are usually complex and sophisticated, the underlying idea of every spectroscopic measurement remains the same: shine a light upon the object of interest or make the object emit the light itself, acquire the light and analyze it. This analysis, which could be conducted with a vast array of elaborate methods, allows one to deepen the knowledge about the nature of matter and light.

From Classical Spectroscopy to Quantum Mechanics

In 1666, Newton decomposed a beam of white light into continuum of colours and named it “spectrum” [1, 2]. What Newton did is often regarded as the first spectroscopic experiment, though not yet a measurement. The first spectroscopic measurement was made in 1814 when Joseph von Fraunhofer detected dark lines in the continuous spectrum of sunlight\(^1\) dispersed by a prism [3]. Later, in 1821, for the same purpose he used a transmission diffraction grating that he had invented. Unlike the prism, the diffraction grating allowed Fraunhofer to calculate the absolute wavelength of the dark stripes from their position in the spectrum. Despite the fact that Fraunhofer did not understand the origin of the black stripes, he used them as a reference for other measurements. The subsequent leap in spectroscopic research was due to John Herschel and W. H. Fox Talbot, who in 1826 reported that each element burnt in the flame gave different pattern of bright lines in the prismatic spectrum. So far, scientists had at their disposal absorption and emission spectroscopy but their relation was still unclear. In 1849, Jean Foucault studied the emission of the electric arc and absorption of sunlight by the same arc. He showed that dark stripe in the spectrum of sunlight transmitted through the arc, corresponding to the \(D\) sodium line, became a bright emission line when the sunlight was shut-off. Combining all the above facts together, Gustav Kirchhoff formulated in 1859 the law of radiation: for a given temperature and wavelength, the ratio of emitted and absorbed power is constant, body-independent, and equals to the black-body emitted power for the

\(^1\)Some of the dark stripes in the spectrum of sunlight were earlier observed by William Hyde Wollaston (in 1802), but they did not draw much of his attention. He gave them the naïve interpretation of being boundaries between adjacent colours.
same temperature and wavelength [4]. One can say that the Kirchhoff law demonstrated spectral equivalence of absorption and emission spectra. In 1860 Kirchhoff, together with Robert Bunsen, published a paper [5] with first systematic analysis of alkali spectra. They proposed a method of a very sensitive determination of chemical composition of materials, emphasizing that temperature does not influence position of the emission lines. This allowed unequivocal assignment a given set of spectral lines to one specific element. Using the same spectroscopic methods, in late 1860, Kirchhoff and Bunsen discovered a new element, cesium, and in early 1861 rubidium which is the element we deal with in this thesis.

The second part of the nineteenth century brought discoveries of myriads of atomic and molecular spectra. However, determination of chemical composition of the analyzed medium was by no means all that could be obtained from spectroscopic observations. The attempts to understand and to explain experimental results required new theoretical approach and soon became the driving horse for the development of quantum mechanics, a theory initiated by Max Planck by his derivation of the law of black-body radiation in late 1900. Constant improvement of the resolution of spectroscopic experiments enabled one to resolve narrower features and to gain deeper insight into internal atomic structure. This resulted in discoveries of new phenomena, such as splitting of atomic energy levels in magnetic and electric fields (Zeeman [6] and Stark effects), fine and hyperfine interactions, etc.

**Laser Spectroscopy**

A giant leap in spectroscopy is due to the advent of laser. In 1917 Albert Einstein predicted stimulated emission [7], which allows amplification of coherent and monochromatic radiation. This concept became the basis of the maser, first built by N. G. Basov, A. M. Prokhorov [8] and C. H. Townes, J. P. Gordon, H. J. Zeiger [9, 10]. The major breakthrough on the way to built a laser was Schawlow’s and Townes’ idea to extend maser action onto optical domain [11]. Shortly afterwards, in 1960, Maiman succeeded in building the first pulsed laser [12]. The history of development of masers and lasers can be found with further references in Ref. [13].

Laser sources soon became a basic research tool for spectroscopy. Precise control of the light parameters allowed deeper studies of the properties of the light-matter interaction more profoundly in the regimes that were not accessible with classical light sources. The advantages of lasers could be presented in countless aspects, but they can be summarized in brief as follows: tunability, monochromacity, coherence, and high intensity. Unlike in the era of classical spectroscopy, when most observations had passive character, lasers gave researchers a possibility to influence the observed medium in a very controlled way and sensitively examine its response [14]. Lasers and laser spectroscopy have already defined many new fields of research [15] and have a great impact on our society [16]. One of the most spectacular achievements of laser spectroscopy is laser cooling and trapping of neutral atoms [17]-[20] which, among other non-trivial applications, allows to examine the quantum behavior of cold, diluted, macroscopic atomic samples organized in periodic structures, the, so-called, optical lattices [21], or subject to Bose-Einstein condensation [22]-[24].
Thesis

The thesis is devoted to spectroscopy of cold rubidium atoms confined in a magneto-optical trap. The atomic sample, whose temperature is as low as 100 µK, is subject to constant interaction with three pairs of intense trapping beams. A probe beam samples the cold atomic cloud at a frequency close to the trapping beam frequency. This nearly degenerate nonlinear process can be a rich source of information about the properties of an atom whose external and internal degrees of freedom are influenced by intense light. In this thesis the attempt is made to interpret and explain the features which appear in the absorption and four-wave mixing spectra obtained in the experiment.

The thesis is organized as follows:

**Chapter 1** concentrates on the quantum semiclassical description of a nonlinear atomic response of a medium perturbed with a strong laser beam (pump) and examined with a weak, non-perturbing probe beam. In the beginning we discuss a theory based on a two-level atom model. This simple model allows one to comprehend basic features of the pump-probe spectroscopy without bothering with the details of multi-level atoms. Since two-level model fails in explanation of many spectral features observed in the experiment, multi-level atom treatment is also introduced in this Chapter.

Influence of the atomic motion on the spectra is presented in **Chapter 2**. The resonant structure of absorption and four-wave mixing signals can be profoundly altered by two contributions: first, an effect due to the localization of atoms in optical micropotentials created by the interference of the trapping beams (optical lattices) and second, involving atoms that travel with small velocities in the trap (the recoil-induced resonances). Although the latter contribution is expected to have greater impact on our results, both are discussed in this chapter.

In **Chapter 3** a brief description of the experimental apparatus is presented. Experimental realizations of magneto-optical trap, laser frequency synchronization, pump-probe spectroscopy and the vacuum system are described.

**Chapter 4** contains the discussion of results obtained in our experiment. Their qualitative and, whenever possible, quantitative analysis is given. In order to do so, we use the theory discussed in Chapters 1 and 2. Also, the conclusions and outlook for further development of our research are presented there.

**Appendices** contain additional remarks, important for the complete picture of the experiment presented in this thesis.
Chapter 1

Atoms in Intense Light Fields

1.1 Introduction

When subject to intense light fields, a medium behaves in a nonlinear way. In other words, the response of a medium is no longer proportional to the amplitude of the incident electric field $E$ and contributions of higher powers of $E$ become significant [25, 26].

Description of the medium response is based on the induced polarization vector $\mathbf{P}$ which is defined as a net dipole moment of the medium per unit volume:

$$\mathbf{P} = \frac{1}{V_n} \sum_{i=1}^{n} \mathbf{d}_i.$$  \hspace{1cm} (1.1)

In formula (1.1), $n$ denotes the number of particles in volume $V_n$ of the medium and $\mathbf{d}_i$ is the dipole moment of the $i$-th particle. We can rewrite formula (1.1) using the particle density $N$ and the average dipole moment $\langle \mathbf{d} \rangle = n^{-1} \sum_{i=1}^{n} \mathbf{d}_i$ and obtain

$$\mathbf{P} = N \langle \mathbf{d} \rangle.$$  \hspace{1cm} (1.2)

The quantum-mechanical description of $\mathbf{P}$ can be introduced in a straightforward manner. If we recall that $\langle \mathbf{d} \rangle = \text{Tr}(\hat{\rho} \hat{\mathbf{d}})$, where $\hat{\rho}$ is a density operator of the system and $\hat{\mathbf{d}}$ is the dipole moment operator, we obtain

$$\mathbf{P} = N \text{Tr} \left( \hat{\rho} \hat{\mathbf{d}} \right).$$  \hspace{1cm} (1.3)

On the other hand, polarization of the medium can be expressed in terms of the electric field vector $\mathbf{E}$ and the electric displacement vector $\mathbf{D} = \epsilon_0 \epsilon_r \mathbf{E}$ [27]

$$\mathbf{P} = \mathbf{D} - \epsilon_0 \mathbf{E} = \epsilon_0 (\epsilon_r - 1) \mathbf{E} = \epsilon_0 \chi_e \mathbf{E},$$  \hspace{1cm} (1.4)

where $\epsilon_0$ is the permittivity of free space, $\epsilon_r$ is the dielectric constant of the medium and $\chi_e$ stands for its susceptibility. From (1.4) follows that $\epsilon_r = 1 + \chi_e$. In a non-magnetic medium (relative permeability $\mu_r = 1$), the complex refraction index $n_r = \sqrt{\epsilon_r}$. Now, recalling that the imaginary part of the refraction index is related to the absorption coefficient $\alpha$,

$$\alpha(\omega) = \frac{2\omega}{c} \text{Im} n_r = \frac{2\omega}{c} \text{Im} \sqrt{1 + \chi_e} \approx \frac{\omega}{c} \text{Im} \chi_e,$$  \hspace{1cm} (1.5)
where $\omega$ is a circular frequency of the incident radiation and $c$ is the speed of light, we can calculate the absorption in the medium. Solving the Schrödinger equation for $\hat{\rho}$, as it will be presented later, we find average $P$ from equation (1.3). Comparing eqs. (1.3) and (1.4) we find $\chi_e$ and subsequently from (1.5) we get $\alpha(\omega)^1$. All the above formulas are valid both for linear and nonlinear case. Indeed, when we return to the equation (1.4) and take into consideration the possible dependence of $\chi_e$ on $E$, we can write $P$ in the form

$$P = \epsilon_0 \chi_e(E) E.$$ (1.6)

This equation shows that if $\chi_e$ is independent of $E$, the response of the medium is proportional to the strength of applied field. However, when the magnitude of the electric field becomes sufficiently large, the dependence of $\chi_e$ on $E$ shows up and the relation between $P$ and $E$ is no longer linear. The most general expression for polarization can be written as

$$P_i(\vec{r}, t) = \epsilon_0 \sum_{\kappa=1}^{(k)} \chi_{i j_1 j_2 ... j_\kappa}(\vec{r}, t) E_j^1(\vec{r}, t) E_j^2(\vec{r}, t) ... E_j^\kappa(\vec{r}, t),$$ (1.7)

where $\chi_{i j_1 j_2 ... j_\kappa}$ is the tensor coefficient of the $(k-1)$-th power of $E$ in the expansion of $\chi_e(E)$ in equation (1.6). Solving equations (1.3) and (1.4) for $\chi_e(E)$ can be performed either non-perturbatively or by perturbative expansion into a power series of $E$.

The $\chi_e$ dependence on $E$ means that light modifies the medium, whose properties in turn influences the medium’s interaction with light. This process is the essence of nonlinear optics.

1.2 Nonlinear response of two-level system

Let us briefly recall after Ref. [25] the outline of the calculation of the density matrix for a two-level system depicted in Fig. 1.1. Two levels, ground $|g\rangle$ and excited $|e\rangle$, are separated by energy $\hbar \omega_0$. The excited level decays at a rate $\Gamma$ to the ground level only, which is assumed to have infinite lifetime. The system is irradiated with a monochromatic wave of the frequency $\omega$. If we denote energy of $|g\rangle$ as $E_g = \hbar \omega_g$, and similarly for $|e\rangle$, the Hamiltonian of the system will be given by

$$\hat{H}_0 = \sum_{i=e,g} E_i |i\rangle \langle i|,$$ (1.8)

while the interaction energy in the dipole approximation can be written as

$$\hat{V}(t) = -\hat{d} \cdot \hat{E}(t).$$ (1.9)

Because the dipole moment is off-diagonal, one can write the non-zero elements of $\hat{V}(t)$ as

$$V_{eg}(t) = \langle e|\hat{V}(t)|g\rangle = -d_{eg} E(t),$$ (1.10)

where $d_{eg} = \langle e|\hat{d}|g\rangle$.

---

1Let us recall the formula (known as the Beer’s law) for the transmitted intensity $I$ over length $l$ through the medium, involving absorption coefficient $\alpha$: $I = I_0 \exp(-\alpha(\omega)l) \approx I_0(1 - \alpha(\omega)l)$ for $\alpha(\omega) \ll 1$.
1.2. NONLINEAR RESPONSE OF TWO-LEVEL SYSTEM

Figure 1.1: Two-level system with resonance frequency \( \omega_0 \) irradiated with light of frequency \( \omega \). Level \( |e\rangle \) decays with rate \( \Gamma \) exclusively to \( |g\rangle \).

The Schrödinger equation for the density operator

\[
\dot{\hat{\rho}} = -\frac{i}{\hbar} \left[ \hat{H}_0 + \hat{V}, \hat{\rho} \right]
\]

(1.11)
yields the following set of differential equations for the elements of the density matrix:

\[
\dot{\rho}_{eg} = -i\omega_0 \rho_{eg} + \frac{i}{\hbar} V_{eg} (\rho_{ee} - \rho_{gg}) ,
\]

(1.12)

\[
\dot{\rho}_{ee} = -\frac{i}{\hbar} (V_{eg} \rho_{ge} - \rho_{eg} V_{ge}) ,
\]

(1.13)

\[
\dot{\rho}_{gg} = -\frac{i}{\hbar} (V_{ge} \rho_{eg} - \rho_{ge} V_{eg}) .
\]

(1.14)

The matrix element \( \rho_{eg} \) is called optical coherence, as it describes the phase relation between wave functions of the ground and excited atomic levels, \( \rho_{gg} \) and \( \rho_{ee} \) are the populations of the ground and excited level, respectively, and \( \omega_0 = (E_e - E_g)/\hbar \). Equations (1.12-1.14), however, do not include the spontaneous processes which result in a radiative decay of the excited level. The appropriate decay terms\(^2\) can be added to obtain

\[
\dot{\rho}_{eg} = -i(\omega_0 + \tilde{\gamma}) \rho_{eg} + \frac{i}{\hbar} V_{eg} (\rho_{ee} - \rho_{gg}) - \tilde{\gamma} \rho_{eg} ,
\]

(1.15)

\[
\dot{\rho}_{ee} = -\frac{i}{\hbar} (V_{eg} \rho_{ge} - \rho_{eg} V_{ge}) - \Gamma \rho_{ee} ,
\]

(1.16)

\[
\dot{\rho}_{gg} = -\frac{i}{\hbar} (V_{ge} \rho_{eg} - \rho_{ge} V_{eg}) + \Gamma \rho_{ee} .
\]

(1.17)

The quantity \( \tilde{\gamma} \) introduced in equation (1.15) is the coherence decay rate and when collisions of atoms are negligible, \( \tilde{\gamma} = \Gamma/2 \) [28], which means that population decays twice as fast as coherence.

We search for the steady-state solution of the density matrix equations of the system perturbed by a monochromatic radiation \( E(t) = E e^{-i\omega t} + c.c. \). Substitution of \( E(t) \) into equation (1.10) and dropping terms which evolve like \( e^{i\omega t} \), leaves us with the set of equations\(^3\)

\[
\dot{\rho}_{eg} = -(i\omega_0 + \tilde{\gamma}) \rho_{eg} + \frac{i}{\hbar} V_{eg} (\rho_{ee} - \rho_{gg}) ,
\]

(1.18)

\[
\dot{\rho}_{inv} = -\Gamma (\rho_{inv} - \rho_{inv}^\text{eq}) - \frac{2i}{\hbar} (V_{eg} \rho_{ge} - \rho_{eg} V_{ge}) ,
\]

(1.19)

\[
V_{eg} = -d_{eg} E e^{-i\omega t} = V_{ge}^* .
\]

\(^2\)The formal justification for this operation is among foundations of the master equation theory [28].

\(^3\)This is part of the rotating wave approximation [29].
where we introduced population inversion \( \rho_{\text{inv}} = \rho_{ee} - \rho_{gg} \). \( \rho_{\text{inv}}^e \) denotes equilibrium value of \( \rho_{\text{inv}} \), i.e., the steady-state population difference in the absence of an incident field, e.g., due to thermal excitation.

To solve the equations we assume that evolution of coherence \( \rho_{eg} \) driven by a monochromatic wave of frequency \( \omega \) can be described as

\[
\rho_{eg}(t) = \sigma_{eg}(t)e^{-i\omega t},
\]

where \( \sigma_{eg}(t) \) is slowly varying envelope of the coherence. After substituting (1.21) into (1.18,1.19)\(^4\), we set all the time derivatives to zero\(^5\) and obtain a set of algebraic equations. Now it is straightforward to obtain the susceptibility \( \chi_e(E) \) from the relation \( P = N \text{Tr} \hat{\rho} \hat{d} = N (d_{ge} \rho_{eg} + \text{c.c.}) = \epsilon_0 \chi_e(E)E \):

\[
\chi_e = \frac{N \rho_{\text{inv}}^e |d_{eg}|^2}{\epsilon_0} \frac{\Delta/\gamma - i}{1 + \Delta^2/\gamma^2 + \Omega^2/\gamma \Gamma},
\]

where \( \Delta = \omega - \omega_0 \) is the detuning of light from atomic resonance and \( \Omega \) is the Rabi frequency, defined as

\[
\Omega = \frac{2|d_{eg}| |E|}{\hbar}.
\]

Indeed, as it can be seen from equations (1.22) and (1.23), \( \chi_e \) is dependent on \( E \).

To examine the contributions of the specific order \( (\kappa - 1) \) to polarization \( P \) of the medium [see (1.4)], \( \chi_e \) should be expanded in series with respect to \( E \) [25]. Rather than doing it, we shall briefly analyze the resulting absorption coefficient \( \alpha \) when \( \gamma = \Gamma/2 \),

\[
\alpha(\Delta) = \frac{\alpha_0}{1 + 2\Omega^2/\Gamma^2} \left(1 + \frac{4\Delta^2}{\Gamma^2(1 + 2\Omega^2/\Gamma^2)}\right)^{-1},
\]

where \( \alpha_0 \) is the on-resonance \( (\Delta = 0) \), unperturbed \( (\Omega/\Gamma \ll 1) \) absorption coefficient given by

\[
\alpha_0 = -2 \frac{N \rho_{\text{inv}}^e |d_{eg}|^2}{\epsilon_0 \hbar \Gamma}.
\]

Figure 1.2: Dependence of the absorption coefficient \( \alpha \) on detuning \( \Delta \) from the atomic resonance for different Rabi frequencies.

---

\(^4\)This completes the rotating wave approximation.

\(^5\)We are interested in steady solution, thereby we assume that in equilibrium the populations and coherence envelopes do not change.
As it can be seen from Figure 1.2, the absorption curve (1.24) decreases and broadens with increase of $\Omega$, hence, the Rabi frequency is a measure of the strength of light-matter interaction. This phenomenon of spectral line power broadening is the most common example of nonlinear response of the medium to intense perturbation.

Let us introduce the dimensionless on-resonance saturation parameter $s_0$, defined as the term in formula (1.24) responsible for line broadening, namely

$$s_0 = \frac{2\Omega^2}{\Gamma^2}.$$  

(1.26)

It can be easily verified that when $s_0 = 1$, the perturbed on-resonance absorption drops to half of its unperturbed value. If we write the saturation parameter as

$$s_0 = \frac{I}{I_{\text{sat}}},$$

(1.27)

we can define saturation intensity $I_{\text{sat}}$ as the intensity which reduces the on-resonance absorption by a factor of 2. Using relation $I = 2(\epsilon_0/\mu_0)^{1/2}|E|^2$ and combining it with (1.23), we can relate Rabi frequency to the intensity of light:

$$\Omega = \sqrt{\frac{2|\Delta g|}{\hbar}} \left(\frac{\mu_0}{\epsilon_0}\right)^{1/4} I^{1/2}.$$ 

(1.28)

Formula (1.28) allows calculation of the saturation intensity if, according to (1.26) and (1.27), we set $\Omega = \Gamma/\sqrt{2}$.

### 1.3 Dressed two-level system

The calculations presented in the previous section show that when registering absorption of the beam tuned close to the resonant frequency $\omega_0$, we expect broadening of the resultant line and a decrease of its amplitude with the increase of the beam intensity.

The phenomenon of power broadening, discussed above, is not the only nonlinear effect occurring when the medium is excited by a strong, nearly resonant beam. In particular, the spectral profile of light spontaneously irradiated by a two-level system demonstrates another familiar example. In order to find main features of such a spectrum, we write the time-dependent Schrödinger equation

$$i\hbar \frac{\partial \psi(\mathbf{r}, t)}{\partial t} = \left(\hat{H}_0 + \hat{V}(t)\right) \psi(\mathbf{r}, t),$$

(1.29)

where $\hat{H}_0$ and $\hat{V}(t)$ are given by eqs. (1.8) and (1.9), respectively, $\mathbf{E}(t)$ in (1.9) is a monochromatic wave of frequency $\omega$ and amplitude $E$, and the wavefunction can be written in the form

$$\psi(\mathbf{r}, t) = \sum_{j=\epsilon,g} C_j(t)u_j(\mathbf{r})e^{-i\omega_j t}. $$

(1.30)

Here, $u_j(\mathbf{r})e^{-i\omega_j t}$ is the wavefunction of atomic state $|j\rangle$ and $C_j(t)$ is the probability amplitude of finding the atom in state $|j\rangle$ at the time $t$. We shall not give the complete mathematical description of this problem here (it can be found, for example, in [25]), we rather concentrate on the analysis of its main signatures.

---

6The width at half maximum is $\delta\omega_{\text{FWHM}} = \sqrt{\Gamma^2 + 2\Omega^2}$.

7The detailed calculations are rather elaborate and rely on the second order dipole correlation functions [30, 31, 32].
On the general form of the solution of (1.29) [25] we impose particular boundary conditions that give such coefficients \( C_j(t) \), whose time dependence explicitly appears only in the phase \( |C_j(t)| = \text{const.} \). The resulting normalized wavefunctions (1.30) can be written in the form [25]

\[
\psi(r,t) = \frac{\omega}{\Omega'} \left[ \frac{\Omega'}{2(\Omega' + \Delta)} \right]^{1/2} u_g(r) \exp \left[ -i(\omega_g - \Omega) t \right] \\
\quad \mp \left[ \frac{(\Omega' + \Delta)}{2\Omega'} \right]^{1/2} u_e(r) \exp \left[ -i(\omega_e + \Omega) t \right],
\]

where \( \Omega' = \sqrt{\Omega^2 + \Delta^2} \) is the generalized Rabi frequency. The states \( \psi(r,t)_\pm \) are called the dressed states [31]-[34]. Let us now have a closer look at their properties. Our requirements imposed on the \( C_j(t) \) coefficients results in a constant admixture of the atomic “unperturbed” state \(|g\rangle\) in any of the dressed states \( \psi(r,t)_\pm \), namely

\[
|\langle g|\psi(r,t)\rangle|^2 = \frac{|\Omega|^2}{2\Omega'(\Omega' + \Delta)} \tag{1.32}
\]

\[
|\langle e|\psi(r,t)\rangle|^2 = \frac{\Omega' + \Delta}{2\Omega'} \tag{1.33}
\]

Now, it is easy to conclude, that in the strong interaction limit, when \( \Omega \gg \Delta \) and \( \Omega' \approx |\Omega| \), both probabilities (1.32,1.33) tend to \( \frac{1}{2} \) and the dressed states become the mixture of \(|g\rangle\) and \(|e\rangle\) levels with equal weights. The opposite situation occurs in a weak interaction limit, \( \Omega \ll \Delta \), and \( \Omega' \approx |\Delta| \). From (1.32,1.33) it follows that for positive detuning \( \Delta \) the main contribution to the state \( \psi(r,t)_+ \) is due to \(|g\rangle\) and \( \psi(r,t)_- \) contains mainly \(|e\rangle\). By reversing sign of \( \Delta \) we arrive at the opposite conclusion: \( \psi(r,t)_+ \) is primarily composed of \(|e\rangle\) and \( \psi(r,t)_- \) is almost entirely influenced by \(|g\rangle\). The dressed states are orthogonal ((\( \langle \psi(r,t)_+ | \psi(r,t)_- \rangle = 0 \)) but the dipole-moment matrix calculated in the base of dressed states has all non-vanishing elements:

\[
\langle \psi(r,t)_+ | \hat{d} | \psi(r,t)_+ \rangle = \pm \frac{\Omega}{2\Omega'} d_{ge} \exp(-i\omega t) + c.c. \tag{1.34}
\]

\[
\langle \psi(r,t)_+ | \hat{d} | \psi(r,t)_- \rangle = \pm \frac{\Omega}{2\Omega'} d_{ge} \left( \frac{\Omega' + \Delta}{\Omega' + \Delta} \right)^{1/2} \exp[-i(\omega + \Omega') t] \\
\quad \mp \frac{\Omega}{2\Omega'} d_{eg} \left( \frac{\Omega' + \Delta}{\Omega' + \Delta} \right)^{1/2} \exp[i(\omega + \Omega') t] \tag{1.35}
\]

These important equations include frequencies of the dipole moment oscillations in the dressed state basis: \( \omega, \omega - \Omega' \) and \( \omega + \Omega' \). When we interpret these oscillations as the result of transitions between appropriate dressed states we can draw a diagram of the new structure of energy levels created by the interaction of light with the atom. The picture of an atom dressed with photons is very convenient for modelling light-shifts of the atomic levels (AC Stark shifts, [34]), for understanding the structure of the spectra of resonance fluorescence [30, 35] and absorption in pump-probe spectroscopy [36].

---

8The other type of solution is characterized by the coefficients \( C_j(t) \) for which \( |C_j(t)| \) oscillates with generalized Rabi frequency \( \Omega' \). Such solution is obtained for an atom initially in the ground (or excited) state and describes the population oscillations.
The dressed states (1.31), although stationary in the sense of constant probabilities (1.32, 1.33), are not eigenstates of energy, because the Hamiltonian in (1.29) is time-dependent. Formally, however, we can derive wavefunctions for dressed states which are eigenfunctions of $\hat{H}$. For this we shall consider the radiation and atom as a whole system and diagonalize the resultant Hamiltonian in the basis of tensor products of atomic states $|i\rangle$ and Fock states $|N\rangle$ [37]. The notation $|i,N\rangle = |i\rangle \otimes |N\rangle$ means the atom in state $|i\rangle$ with energy $\hbar \omega_i$ together with $N$ photons of frequency $\omega$. The energy of the state $|i\rangle \otimes |N\rangle$ is $\hbar (\omega_i + \omega)$. The matrix representation of the full Hamiltonian including atom-photon interaction in the basis $|g,1\rangle$ and $|e,0\rangle$ can be written as

$$\hat{H} = \hbar \begin{pmatrix} \omega & -\frac{1}{2} \Omega \\ -\frac{1}{2} \Omega & \omega_0 \end{pmatrix},$$

(1.36)

where we have chosen the zero of the energy scale to coincide with the energy of $|g\rangle$ such also that $\hat{H}_0|g, 1\rangle = \hbar \omega$ and $\hat{H}_0|e, 0\rangle = \hbar \omega_0$, $\hat{H}_0$ is the free Hamiltonian of the atom and the laser field, and phases of the dipole moment and electric field are chosen so that $\Omega$ is real (and positive). The eigenvalues of $\hat{H}$ are

$$\varepsilon_\pm = \frac{\hbar}{2} (\omega + \omega_0 \mp \Omega'),$$

(1.37)

with the corresponding eigenstates

$$|\pm\rangle = \frac{\Omega}{(2\Omega'(\Omega' \mp \Delta))^{1/2}} |g, 1\rangle \mp \left(\frac{\Omega' \mp \Delta}{2\Omega'}\right)^{1/2} |e, 0\rangle.$$  

(1.38)
Figure 1.4: Energies of states of a two-level system modified by light in case of (a) strong perturbation ($\Omega \gg \Gamma$) and (b) weak perturbation ($\Omega \ll \Gamma$). The thickness of grey lines represents the width of the levels. Note that in case of resonance ($\Delta = 0$) and strong perturbation, the perturbed levels are split by the amount of $\Omega'$, contrary to the case of weak perturbation when they cross. In the limit of $\Delta \to \pm \infty$, the perturbed states asymptotically approach relevant $|i,N\rangle$ states (no atom-field coupling). The departure of the perturbed states from the asymptotes are the AC Stark shifts of atomic levels. The plot (b) was obtained from the expansion of the generalized complex Rabi frequency $\Omega' = \sqrt{\Omega^2 + (\Delta - i\Gamma/2)^2}$ for $\Omega \ll |\Delta|, \Gamma$ (see Appendix A).
Thus, the dressed states can be expressed as linear combinations of states of an atom accompanied by appropriate number of photons\(^9\). The atom-photon interaction, whose strength is measured by the Rabi frequency \(\Omega\), creates a mixture of the \(|i, N\rangle\) states. Their properties, such as populations and decay rates\(^10\), are transferred to the resultant \(|\pm\rangle\) states with contributions governed by the relevant coefficients in (1.38). Although we considered the ground state accompanied by one photon and excited state with no photons, the calculations are valid for a general case of \(|g, N + 1\rangle\) and \(|e, N\rangle\) states. The dressed states which are linear combinations of \(|g, N + 1\rangle\) and \(|e, N\rangle\) can be treated as a manifold indexed by the number of photons \(N\), namely \(|\pm\rangle(N)\). Hence, the dressed atom appears as a periodic ladder of manifolds \(|\pm\rangle(N)\) separated by the energy \(\hbar \omega\). The relative energies of the dressed components \(|\pm\rangle(N)\) of manifold \((N)\) are shown in Figure 1.4.

As it was mentioned before, the picture of the dressed atom can be used to explain the spectra of resonance fluorescence. We can now understand this process as a radiative cascade down the ladder of manifolds of dressed states [37], which occurs at the three frequencies of a familiar Mollow triplet, \(\omega\) and \(\omega \pm \Omega'\).

1.4 Pump-probe absorption spectroscopy

Until now we considered a medium which interacted with only one intense monochromatic beam. The beam frequency \(\omega\) was tuned around the resonance frequency \(\omega_0\) of the system and we discussed its absorption (eq. 1.24, Figure 1.2). Now we shall focus on the case when the medium, saturated with an intense beam of a fixed frequency \(\omega\) is probed by a weak beam of intensity well below the saturation, which is tuned around \(\omega\) by small amount \(\delta\). We record absorption of the probe beam in the saturated medium as a function of \(\delta\) (Figure 1.5). The intense beam which saturates atomic transition is called “pump beam”, thus the spectroscopic method presented above is called “pump-probe absorption spectroscopy”. Because the pump beam dresses atomic states with photons and the weak beam probes such a system, we can predict positions of the absorption resonances directly from the picture of dressed atom.

We expect resonances to occur when the probe frequency hits one of the frequencies \(\omega\) or \(\omega \pm \Omega'\). In order to predict the sign of the absorption coefficient connected with these transitions, let us assume that the pump beam detuning \(\Delta = \omega - \omega_0\) is negative. From equations (1.32, 1.33) we know that the upper dressed state \(|\psi_+\rangle\) (or \(|+\rangle\) in the tensor basis \(|i, N\rangle\)) consists mainly of \(|e\rangle\) and lower state \(|\psi_-\rangle\) \((|-\rangle)\) mainly of \(|g\rangle\), hence population of state \(|\psi_-\rangle\) is greater than of \(|\psi_+\rangle\) (as it is depicted in Figure 1.3). Transition at frequency \(\omega + \Omega\) \((\delta = \Omega)\) connects the lower, more populated state with the higher, less populated state. Consequently, absorption overcomes stimulated emission and probe beam is thereby attenuated. On the contrary, the transition at frequency \(\omega - \Omega\) connects the states that show population inversion. It results in stimulated emission of photons to the probe beam, hence probe gain is observed (see Figure 1.6). Using the dressed atom picture, we cannot,

\(^9\)Brief inspection shows that states (1.38) have analogous form as (1.31).

\(^{10}\)Although we did not consider the decay rates of atomic levels, it is straightforward to introduce them by replacing the real atomic frequency \(\omega\) of the state \(|i\rangle\) by the complex one, namely \(\omega'_i = \omega_i - i \Gamma_i/2\). Imaginary part of the resulting complex energy (1.37) which incorporates complex \(\Omega'\) is the width of the corresponding state (see Figure (1.4)).

\(^{11}\)They correspond to probe-pump detuning \(\delta = 0, \pm \Omega\), respectively.
Figure 1.5: The idea of pump-probe absorption spectroscopy. Intense pump beam of fixed frequency $\omega$ is passed through the medium characterized by resonant frequency $\omega_0$ and excited level decay rate $\Gamma$. A weak probe beam ($I_{\text{probe}} \ll I_{\text{sat}}$) is then introduced and tuned around the pump beam frequency by small amount $\delta$. Absorption of the probe beam is monitored by the photodetector. Whilst the pump beam experiences absorption described by equation (1.24), the probe beam samples the structure of the medium dressed by the pump beam.

however, predict what happens to the probe when its frequency is tuned to $\omega$, as populations of the levels connected by this transition are equal.

Detailed calculations of the pump-probe spectra for the two-level system are presented in [38, 39, 40, 25]. Their main concept is to treat the pump field $E_0$ to all orders of magnitude and the probe field $E_1$ only to the first order. We solve density matrix equations (1.18, 1.19) with interaction energy in a rotating wave approximation

$$V_{eg} = -d_{eg} (E_0 e^{-i\omega t} + E_1 e^{-i(\omega + \delta)t}) = V_{ge}^*, \quad (1.39)$$

remembering that $|E_1| \ll |E_0|$. Keeping in mind this approximation, we expect the steady state population inversion $\rho_{\text{inv}}$ and coherence $\sigma_{eg}$ to be of the form

$$\rho_{\text{inv}} = \rho_{\text{inv}}^{(0)} + \rho_{\text{inv}}^{(1)} e^{-i\delta t} + \rho_{\text{inv}}^{(-1)} e^{i\delta t}, \quad (1.40)$$

$$\sigma_{eg} = \sigma_{eg}^{(0)} + \sigma_{eg}^{(1)} e^{-i\delta t} + \sigma_{eg}^{(-1)} e^{i\delta t}, \quad (1.41)$$

where $\rho_{eg} = \sigma_{eg} e^{-i\omega t}$ [compare (1.21)]. After introducing these expressions to the density matrix equations, dropping all the terms which are proportional to the second and higher powers of the quantities with index $(\pm 1)$ and comparing the terms oscillating at the same frequencies, we arrive at a set of algebraic equations which are then solved for the variables $\rho_{\text{inv}}^{(0, \pm 1)}$ and $\sigma_{eg}^{(0, \pm 1)}$. We are interested in $\sigma_{eg}^{(1)}$, the spectral component of $\rho_{eg}$ which is associated with the probe frequency $\omega_{pr} = \omega + \delta$. The imaginary part of $\sigma_{eg}^{(1)}$ is proportional to the absorption coefficient experienced by the probe beam [see equation (1.5)], thus it governs the shape of the signal registered

Figure 1.6: Spectrum of the probe beam tuned by $\delta$ around the frequency of the pump beam $\omega$, predicted from the picture of dressed atom for the pump detuning $\Delta < 0$. 
by photodetector. We obtain [25]

\[
\sigma_{eg}^{(1)} = \frac{d_{eg} \rho_{\text{inv}}^{(0)}}{D(\delta)} \left[ (\delta + i\Gamma) \left( \delta - \Delta + i\Gamma/2 \right) - \frac{\Omega^2}{2} \frac{\delta}{\Delta - i\Gamma/2} \right] E_1, \tag{1.42}
\]

where \( \rho_{\text{inv}}^{(0)} \) is steady-state population inversion introduced by the pump beam alone,

\[
\rho_{\text{inv}}^{(0)} = \rho_{\text{eq}}^{\text{inv}} \frac{1 + 4\Delta^2/\Gamma}{1 + 4\Delta^2/\Gamma + 2\Omega^2/\Gamma}, \tag{1.43}
\]

and \( D(\delta) \) is defined as

\[
D(\delta) = \delta \left( \delta^2 - \Omega^2 - \frac{5\Gamma^2}{4} \right) + i\frac{\Gamma}{2} \left( 4\delta^2 - 2\Delta^2 - \Omega^2 - \frac{\Gamma^2}{2} \right). \tag{1.44}
\]

Although formula (1.42) together with substitutions (1.43,1.44) is somewhat complicated, it is feasible to analyze its resonant structure governed by generally complex denominator \( D(\delta) \) (1.44). Looking at the poles of (1.42), we see that in the limit of strong interaction with the pump \((\Omega \Gamma/2 \gg 1)\), resonances of (1.42) occur when the probe-pump detuning \( \delta \) equals 0, \( \pm \Omega' \). By inspection of the imaginary part of (1.44) for \( \delta = 0, \pm \Omega' \) we can deduce the width of relevant resonances, namely

\[
\Gamma_{\delta=0} = \frac{\Gamma}{2} \left( 1 + \frac{\Delta^2}{\Omega^2 + \Delta^2} \right), \tag{1.45}
\]

\[
\Gamma_{\delta=\pm \Omega'} = \frac{\Gamma}{2} \left( 1 + \frac{\Omega^2}{2(\Omega^2 + \Delta^2)} \right). \tag{1.46}
\]

As we can see, the solutions that we obtained from the density matrix equations coincide with the predictions derived from the picture of the dressed atom on page 13. The imaginary part of (1.42) which is proportional to the absorption coefficient was plotted in Figure 1.7 for \( \Delta = -3\Gamma \) and \( \Omega = 10\Gamma \). The signs of resonances (gain, absorption) for relevant \( \delta = 0, \pm \Omega' \) also agree with the dressed atom model predictions. Moreover, the calculations properly predict the shape of the central feature which was impossible within the dressed atom model.

### 1.5 Four-wave mixing in two-level system

In the preceding section we concentrated on the density matrix element \( \sigma_{eg}^{(1)} \) (1.42), oscillating at frequency \( \omega + \delta \) of the probe beam, thus responsible for the probe beam absorption. There is, however, another spectral component \( \sigma_{eg}^{(-1)} \) of \( \rho_{eg} \) oscillating at frequency \( \omega - \delta \), resulting from the first-order in \( E_1 \) treatment of the pump-probe density matrix equations [see (1.41)]. It is given by

\[
\sigma_{eg}^{(-1)} = \frac{2\rho_{\text{inv}}^{(0)} d_{eg} |d_{ge}|^2 \left( \delta - \Delta - i\frac{\Gamma}{2} \right) \left( -\delta + i\Gamma \right)}{\hbar^3 \left( \Delta - \delta + i\frac{\Gamma}{2} \right) \left( \Delta + i\frac{\Gamma}{2} \right) D^*(\delta)} E_0^2 E_1^*. \tag{1.47}
\]

We see that \( \sigma_{eg}^{(-1)} \) is directly proportional\(^\text{12}\) to \( E_0^2 E_1^* \), unlike \( \sigma_{eg}^{(1)} \) which is explicitly proportional only to \( E_1 \). Thus, by the convention introduced in Ref. [25], we can

\(^{12}\)By direct proportionality we mean the situation when the corresponding electric field amplitude \( E_{0,\pm 1} \) appears directly in a given term. We shall keep in mind, however, that \( \Omega \), which also involves \( E_0 \), appears in density matrix equations in a nonlinear fashion.
write the amplitude of polarization of the medium as the sum of two components, one oscillating at the probe frequency $\omega+\delta$ and the other oscillating at the frequency $\omega-\delta$, namely

$$p(t) = \chi^{\text{ABS}}_{\text{eff}}(\omega + \delta) E_1 e^{-i\delta t} + 3 \chi^{\text{FWM}}_{\text{eff}}(\omega - \delta) E_0^2 E_1^* e^{i\delta t},$$  

where $P(t) = p(t) e^{-i\omega t} + c.c.$ and effective susceptibilities are given by equations

$$\chi^{\text{ABS}}_{\text{eff}}(\omega + \delta) = \sigma^{(1)}_{eg} d_{ge} (\hbar E_1)^{-1}$$  

$$\chi^{\text{FWM}}_{\text{eff}}(\omega - \delta) = \sigma^{(-1)}_{eg} d_{ge} (3\hbar E_0^2 E_1^*)^{-1}.$$ 

Figure 1.8: Pump-probe spectroscopy where generation of the fourth wave (depicted by the dashed arrow) of frequency $\omega-\delta$ was taken into consideration.
as a source of a new wave, generated in the nonlinear process called “four-wave mixing” (FWM). The name originates from the fact that the process involves two photons from the pump field, one from the probe, and the fourth photon is the generated one. Thus, we shall supplement the scheme of pump-probe experiment (Figure 1.5) by the generated fourth wave. This is done in Figure 1.8. If we were able to isolate the fourth wave from the probe beam and register its intensity as the function of probe-pump detuning $\delta$, we would obtain the signal of amplitude proportional to $|\sigma_{eg}^{(-1)}|^2$. The FWM signal is plotted in Figure 1.9 for the same parameters as the absorption signal depicted in Figure 1.7.

![Four-wave mixing signal](image)

Figure 1.9: Four-wave mixing signal, proportional to $|\sigma_{eg}^{(-1)}|^2$, plotted for $\Delta = -3\Gamma$ and $\Omega = 10\Gamma$ using formula from Ref. [40]. The fourth wave is generated only for appropriate $\delta$, hence the signal appears on zero background. Resonances occur at the same frequencies as in the case of absorption signal depicted in Figure 1.7.

### 1.6 Phase-matching condition in four-wave mixing

The source term responsible for generation of the fourth wave at frequency $\omega - \delta$ is proportional to $E_0^2 E_1^*$, where $E_0$ and $E_1$ are complex amplitudes of the pump and probe beams, respectively. We recall that the total optical field interacting with medium is given by the formula [compare with (1.39)]

$$E_{\text{total}}(t) = E_0 e^{-i\omega t} + E_1 e^{-i(\omega + \delta)t} + c.c. \quad (1.51)$$

By taking into account the spatial dependence of $E_0$ and $E_1$ one can determine the direction of propagation of the fourth wave. Let us approximate the pump and probe beams by plane waves with wavevectors $\mathbf{k}$ and $\mathbf{k}_{\text{pr}}$, respectively. By writing $E_0(r) = A_0 \exp(ik \cdot r)$ and $E_1(r) = A_1 \exp(ik_{\text{pr}} \cdot r)$, where $A_0$, $A_1$ denote complex amplitudes of the corresponding waves, we can deduce the form of the fourth wave, namely

$$E_{4th}(r, t) \propto E_0(r)^2 E_1^*(r)e^{-i(\omega - \delta)t} = A_0^2 A_1^* e^{-i((\omega - \delta)t - (\mathbf{k} + \mathbf{k}_{\text{pr}}) \cdot \mathbf{r})}. \quad (1.52)$$

---

**This will be discussed in the following sections.**
From equation (1.52) it follows that the fourth wave propagates in the direction of the wave vector

\[ \mathbf{k}_{4\text{th}} = \mathbf{k} + \mathbf{k} - \mathbf{k}_{pr}. \]  

(1.53)

This equation is often called the “phase-matching condition” since the propagation of the generated signal by (1.53) is associated with phase-matching of all incident optical fields.

Now, assuming that both pump and probe beams are co-linear and their wave vectors are parallel to \( z \)-axis, we obtain that the fourth wave also propagates along \( z \)-axis, namely \( \mathbf{k}_{4\text{th}} = k\hat{z} \), where \( \hat{z} \) is the \( z \)-axis unit vector and \( k = |\mathbf{k}| \). The consideration presented above fully justifies the way we drew the fourth wave in Figure 1.8.

![Figure 1.10: Four-wave mixing process in a two-level atom. Two photons (grey arrows, frequency \( \omega \)) from the pump beam are absorbed and one photon is emitted to the probe beam (continuous arrow, frequency \( \omega + \delta \)). As a result, the fourth photon is generated (dashed arrow, frequency \( \omega - \delta \)). Note that the system returns to its initial state.](image)

Looking at equation (1.52) we can interpret the four-wave mixing process as a result of absorption of two pump photons and emission of one photon to the probe beam, as it is depicted in Figure 1.10. Due to the generation of the fourth wave, this nonlinear interaction leaves the atom in its initial state. The coincidence of the virtual levels denoted by the dashed lines with the real or dressed atomic levels significantly increases the probability of the process.

Now, let us consider a different experimental situation in which we use two counter-propagating pump beams of the same frequency \( \omega \) and the wave vectors \( \mathbf{k}_f \) and \( \mathbf{k}_b = -\mathbf{k}_f \) along \( z \)-axis. We probe the medium with a weak beam of frequency \( \omega + \delta \) sent at a small angle with respect to one of the pump beams (Figure 1.11).

As before, the generation of the fourth wave can occur by absorbing two photons from one of the pump beams and emitting one photon to the probe beam. There is, however, yet another possibility. Two pump photons are absorbed, but each from the different pump beam. In such a case, because \( \mathbf{k}_f = -\mathbf{k}_b \), the fourth wave propagates in opposite direction to the probe beam, since \( \mathbf{k}_{4\text{th}} = \mathbf{k}_f + \mathbf{k}_b - \mathbf{k}_{pr} = -\mathbf{k}_{pr} \). Setting appropriately two detectors, we can simultaneously measure absorption of the probe beam and the four-wave mixing signal. Such a setup was used in the experiment presented in this thesis.

\[ ^{14} \text{In deriving the expression for } \mathbf{k}_{4\text{th}} \text{ we utilized the fact that for } \delta/\omega \ll 1, |\mathbf{k}_{pr}| \cong |\mathbf{k}| = k. \]
1.7 Grating picture of four-wave mixing

Let us assume four-wave mixing with both pump beams and the probe beam of the same frequency $\omega$. This particular case is often called the degenerate four-wave mixing (DFWM). Now, consider the interference structure produced by the probe beam and forward pump beam, $k_f$. After simple calculations we obtain that these beams create periodic light intensity modulation with the period

$$d_f = \frac{\lambda}{2 \sin \frac{\theta}{2}},$$

(1.54)

where $\lambda$ is the wavelength of incident optical fields and $\theta$ is the angle between probe and pump beams. This modulation can be depicted as the planes of maximum intensity making an angle $\frac{\theta}{2}$ with the pump beams’ propagation axis. Such a modulation of intensity implies periodic modulation of the medium properties, creating in it a grating with period $d_f$. This can be a population grating in the case of a two-level atom, or a coherence grating when the multi-level atom and appropriate polarizations of laser beams are considered [41]. The other pump beam, $k_b$, is diffracted in a Bragg-like fashion from the grating. It can be easily verified that the direction of diffraction is exactly opposite to the direction of probe beam [see Figure 1.12(a)], since the Bragg condition coincides with phase-matching condition (1.53).

Analogous consideration should apply to another pair of the pump-probe beams, namely $k_b$ and $k_{pr}$. In this case, the interference pattern is characterized by the period

$$d_b = \frac{\lambda}{2 \cos \frac{\theta}{2}},$$

(1.55)

and makes an angle $90^\circ + \frac{\theta}{2}$ with the pump beams axis. Now, the forward pump $k_f$ is also diffracted by the grating oppositely to the probe beam propagation [see Figure 1.12(b)]. The fourth wave in the grating picture consists of photons of both pump
beams, diffracted off the gratings created by periodic modulation of the medium properties by the probe and pump beams.

![Diagram of four-wave mixing process](image)

Figure 1.12: The four-wave mixing process viewed as composed by two contributions: (a) diffraction of backward pump beam \( k_b \) on a grating induced by interference of probe beam \( k_{pr} \) and forward pump beam \( k_f \) (grating period \( d_f \)) and (b) diffraction of the forward pump beam on a grating induced by interference of the probe beam and the backward pump beam (grating period \( d_b \)). The beams responsible for grating creation are depicted as black arrows, beams undergoing diffraction appear as grey, bold arrows.

### 1.8 Nonlinear optics in two-level system - summary

In the preceding sections we have presented principles of nonlinear processes of interest that occur in the two-level system. We have shown that interaction of light with a resonant medium can dramatically change its properties which, in turn, alters the response of the medium to incident radiation. As the most basic example we considered saturation of absorption and spectral line broadening (Section 1.2). Next we concentrated on studying a medium perturbed by an intense pump beam of fixed frequency with a weak, tunable probe beam (Section 1.4). We used the dressed atom approach to explain the shape of the spectrum obtained when the probe beam is tuned around the frequency of the pump beam (Section 1.3). The nonlinear effects in such a system manifest themselves in vanishing of the resonance at the atomic transition frequency and appearance of three resonances, whose positions and signs (gain or absorption) are determined by the quantities: Rabi frequency \( \Omega \) and pump beam detuning from atomic resonance \( \Delta \). These quantities characterize the atom-field interaction. Moreover, the dressed-atom picture can be used to understand the origin of the AC Stark shifts of atomic levels. Finally, as a consequence of nonlinearity in the atom-field interaction we demonstrated the possibility of generating coherent radiation in the four-wave mixing process and analyzed its spectral composition (Section 1.5).

The calculation of either absorption or the four-wave mixing signal aims at finding the spectral component of polarization \( \mathbf{P} \) of the medium oscillating at appropriate frequency. This, in turn, amounts to calculation of the density matrix \( \hat{\rho} \), which yields the expectation value of the dipole operator \( \hat{d} \). Since \( \hat{d} \) has only off-diagonal non-zero elements and is determined exclusively by an internal atomic structure,
the absorption and four-wave mixing signals are governed by the off-diagonal density matrix elements $\rho_{eg}$. The first is proportional to $\text{Im} [\rho_{eg}(\omega + \delta)]$ and the latter to $|\rho_{eg}(\omega - \delta)|^2$. For pure states there is an equivalent approach, which consists in averaging $\hat{d}$ operator using wavefunctions of appropriate states, as it was done for the dressed atom model.

The experiment discussed in this thesis uses methodology of the pump-probe absorption spectroscopy and four-wave mixing with counter-propagating pump beams described above. Although the medium under examination consists of multilevel atoms, the conclusions resulting from the two-level model are important as the initial step in explaining the observations. As it will be shown later, some of the features of the observed spectra can be qualitatively understood in terms of the two level approach. Moreover, the two-level model offers a relatively easy way to gain insight into the light shifts of the levels of multi-level system when appropriate light polarization allows expansion into individual two-level subsystems (see Section 1.13). All this justifies the amount of attention that we devoted to the two-level system and its interaction with light.

1.9 Degenerate two-level system (DTLS)

In the following sections we will consider the degenerate two level system (DTLS) perturbed by a strong pump beam and probed with a weak probe beam. The structure of the DTLS is comprised of two levels, the ground level with total angular momentum $F_g$ and the upper (excited) level with total angular momentum $F_e$. The ground and upper levels of DTLS are the manifolds consisting of $2F_g + 1$ and $2F_e + 1$ Zeeman sublevels, respectively, and are separated by the resonant frequency $\omega_0$. In the absence of perturbation, all Zeeman sublevels of the given state have the same energy, which explains the origin of “degeneracy” in the name of DTLS. This degeneracy can be removed by introducing an external perturbation, which could be a magnetic field $\mathbf{B}$ or laser light.

Figure 1.13: The $F_g = 3 \rightarrow F_e = 4$ transition with appropriate Clebsch-Gordan coefficients (in circles) and relative line strengths (numbers by the transition lines). The relative line strengths are defined in units of the weakest transition line strength $m_g = \pm 3 \leftrightarrow m_e = \pm 2$.
In this thesis we investigate one of the components of the $^{85}\text{Rb} \, D_2$ line, the $^5S_{1/2}(F_g = 3) \leftrightarrow ^5P_{3/2}(F_e = 4)$ transition. The DTLS associated with this transition is depicted in Figure 1.13 together with the Clebsch-Gordan coefficients and relative line strengths corresponding to the transitions between the appropriate Zeeman sublevels. Transitions between given sublevels of the ground and excited levels are allowed only for certain polarizations of the incident laser field. To establish a consistent description of this situation we shall now define polarizations of the laser beams with respect to the geometry of the experimental setup.

Most often we choose the quantization axis either parallel to the polarization of a linearly polarized beam or along the direction of propagation of one of the laser beams, as it is done in Figure 1.14. Here, the quantization axis is parallel to $z$ axis. Let us assume that the laser beam propagating along $z$ is circularly polarized. We say that the beam has polarization $\sigma^+$ ($\sigma^-$) when its $E$ vector rotates counterclockwise (clockwise) in the $xy$ plane. The $\sigma^\pm$-polarized beam induces transitions between levels $m_g \leftrightarrow m_e = m_g \pm 1$, which is shown in Figure 1.14 for the simple case of the $F_g = 0 - F_e = 1$ DTLS. Now, consider another beam which is sent perpendicularly to the first one. We define $\pi$ polarization to be parallel to the quantization axis. The transitions allowed for the $\pi$-polarized beam occur between levels with $m_g \leftrightarrow m_e = m_g$. Finally, when polarization of the beam is perpendicular to the quantization axis, the transitions $m_g \leftrightarrow m_e = m_g - 1$ and $m_g \leftrightarrow m_e = m_g + 1$ occur simultaneously. We call such polarization $\sigma$. Additionally, in the case when the beam along $z$ is linearly polarized, we decompose its polarization vector into two circular polarizations, $\sigma^+$ and $\sigma^-$ with their relative phase governed by the angle which the polarization plane makes with the $xz$ plane.

![Figure 1.14: Definitions of polarizations of the incident laser beams with respect to the geometry of the experiment. The quantization axis is taken along $z$ axis which coincides with the direction of propagation of the circularly polarized beam. For given polarizations, the allowed transitions between sublevels of the $F_g = 0 - F_e = 1$ DTLS are shown. The beam propagating along $z$ can be circularly polarized, which is characterized by polarizations $\sigma^+$ and $\sigma^-$, or by linear polarization $\sigma$. The beam propagating along $x$ can be polarized linearly ($\pi$ or $\sigma$). If such beam is circularly polarized, it can always be decomposed into two linear components with a 90° phase-shift between them.](image-url)
1.10 Density matrix description of the DTLS

To calculate the response of the DTLS in the presence of pump and probe laser fields, we use similar approach to that presented in Section 1.4 on page 14. First, we will solve the master equation for the DTLS [28] in the presence of the pump field only and obtain the exact stationary solution for the density matrix\(^{15}\). Next, we use this solution to determine response of the DTLS to the probe beam which will be considered only to the first order of magnitude. We shall, however, keep in mind that such a treatment is valid only in the regime when \( |E_1| \ll |E_0| \), \( |E_1| \ll E\text{sat} \), where \( E_0, E_1 \) are the amplitudes of the pump and probe fields, respectively, and \( E\text{sat} \) is the amplitude of the field that corresponds to the saturation intensity defined in Section 1.2.

This time, however, we have to consider all sublevels of the ground state, \( |F_g, m_g\rangle \) \((m_g = -F_g, \ldots, F_g)\), and excited state \( |F_e, m_e\rangle \) \((m_e = -F_e, \ldots, F_e)\), which increases the Hilbert space dimension to \( 2(F_g + F_e) + 2 \). The density matrix has now \( (2(F_g + F_e) + 2)^2 \) elements:

\[
\hat{\rho} = \begin{pmatrix}
\rho_{-F_g,-F_e} & \cdots & \cdots & \rho_{-F_e,F_g} \\
\vdots & \ddots & \ddots & \vdots \\
\vdots & \ddots & \ddots & \vdots \\
\rho_{F_g,-F_e} & \cdots & \cdots & \rho_{F_e,F_g}
\end{pmatrix}
\begin{pmatrix}
\rho_{-F_g,-F_e} & \cdots & \cdots & \rho_{-F_e,F_g} \\
\vdots & \ddots & \ddots & \vdots \\
\vdots & \ddots & \ddots & \vdots \\
\rho_{F_g,-F_e} & \cdots & \cdots & \rho_{F_e,F_g}
\end{pmatrix}
\]

(1.56)

The diagonal elements of \( \hat{\rho} \) are the populations of appropriate states. The elements with indices belonging to different manifolds, \( F_g \) and \( F_e \), are the optical coherences, for example \( \rho_{F_e,F_g} \) denotes the coherence between states \( |F_g, m_g = F_g\rangle \) and \( |F_e, m_e = F_e\rangle \). Finally, the elements with different indices but of the same manifold, are the so-called Zeeman coherences, i.e. coherences between sublevels of the same manifold. For example, \( \rho_{F_g,-1,F_g} \) stands for the coherence between \( |F_g, m_g = F_g - 1\rangle \) and \( |F_g, m_g = F_g\rangle \) sublevels of the ground state \( F_g \). For the sake of compactness we introduce the projection operators

\[
\hat{P}_g = \sum_{m_g = -F_g}^{F_g} |F_g, m_g\rangle \langle F_g, m_g|,
\]

(1.57)

\[
\hat{P}_e = \sum_{m_e = -F_e}^{F_e} |F_e, m_e\rangle \langle F_e, m_e|.
\]

(1.58)

Since in the absence of external perturbation, all sublevels of the ground (excited) level \( F_g \) \((F_e)\) of DTLS have the same energy \( E_g \) \((E_e)\), we can write the free Hamiltonian of the system in the form

\[
\hat{H}_0 = E_g \hat{P}_g + E_e \hat{P}_e.
\]

(1.59)

The interaction part \( \hat{V} \) of the complete Hamiltonian \( \hat{H} = \hat{H}_0 + \hat{V} \) is given by the following equation (we drop the spatial phase factor \( e^{i\mathbf{k}\cdot\mathbf{r}} \))

\[
\hat{V} = \hat{V}^{(0)} + \hat{V}^{(1)} = -E_0 e^{-i\omega t} \hat{\epsilon}_0 \cdot \hat{d} - E_1 e^{-i\omega t} \hat{\epsilon}_1 \cdot \hat{d} + \text{c.c.},
\]

(1.60)

\(^{15}\)It amounts to considering the pump field to all orders of magnitude.
where \( \varepsilon_0 \) and \( \varepsilon_1 \) are the polarization unit vectors of the pump and probe beams, respectively.

The master equation for density matrix of the DTLS can be written in the form [28, 42]:

\[
\frac{\partial \hat{\rho}}{\partial t} = -\frac{i}{\hbar} \left[ \hat{H}_0 + \hat{V}, \hat{\rho} \right] - \frac{\Gamma}{2} \{ \hat{P}_e, \hat{\rho} \} + \sum_{q=-1,0,1} \hat{Q}^{(q)}_{ge} \hat{\rho} \hat{Q}^{(q)}_{eg} - \gamma (\rho - \rho_0),
\] (1.61)

where the curly brace \{ \ldots \} denotes anti-commutator and \( \hat{Q}^{(q)}_{ge} \) is the \( q \)-th component of vector

\[
\hat{Q}_{ge} = \sqrt{2F_e+1} \frac{\hat{P}_g \hat{d} \hat{P}_e}{\langle F_g || \hat{d} || F_e \rangle},
\] (1.62)

in the spherical basis (see, for example, Ref. [43]). \( \langle F_g || \hat{d} || F_e \rangle \) is the reduced matrix element of the dipole operator \( \hat{d} \). The first term in equation (1.61) describes evolution of the DTLS subject to external perturbation \( \hat{V} \) which in our case is the pump-probe laser field. The second term in (1.61) corresponds to spontaneous relaxation of the excited state populations \( \rho_{m_e,m_e} \) and Zeeman coherences \( \rho_{m_e,m_e'} \), \( m_e \neq m_e' \) with the rate \( \Gamma \) and decay of optical coherences \( \rho_{m_g,m_g} \) with the rate \( \gamma/2 \). The third term is responsible for feeding the ground state populations and Zeeman coherences by spontaneous emission. Indeed, when we substitute (1.62) into the third term of (1.61) and use the Wigner-Eckart theorem

\[
\langle F_e, m_e | \hat{d}^{(q)} | F_g, m_g \rangle = \frac{1}{\sqrt{2F_e+1}} \langle F_e, m_e | F_g, 1, m_g, q \rangle \langle F_e || \hat{d} || F_g \rangle,
\] (1.63)

we obtain the non-vanishing matrix elements of the corresponding operator

\[
\langle F_g, m_g | \hat{Q}^{(q)}_{ge} \hat{\rho} \hat{Q}^{(q)}_{eg} | F_g, m_g' \rangle =
\langle F_e, m_g + q | F_g, 1, m_g, q \rangle \langle F_e, m_g' + q | F_g, 1, m_g', q \rangle \rho_{m_e=m_g+q,m_e'=m_g'+q}.
\] (1.64)

\( \langle F_e, m_e | F_g, 1, m_g, q \rangle \) denote the Clebsch-Gordan coefficients and \( q \) defines polarization of the photon involved in the transition between these levels (\( q = \pm 1,0 \) refers to \( \sigma^+,\sigma^\pi \) respectively). Finally, the last term in equation (1.61) refers to a finite time of interaction of atoms with the laser field which is of the order of \( \gamma^{-1} \). The \( \hat{\rho}^{(0)} \) is the thermal equilibrium density matrix which, as it can be easily verified, is the stationary solution of the master equation (1.61) in the absence of laser perturbation (with \( \hat{V} = 0 \)). We assume that in such a situation the only non-zero elements of the density matrix correspond to the isotropically populated sublevels of the ground manifold, hence

\[
\hat{\rho}^{(0)} = \frac{\hat{P}_g}{2F_g + 1}.
\] (1.65)

### 1.11 Solution of the master equation for DTLS

Let us define the following operators

\[
\hat{\rho}_{ee} = \hat{P}_e \hat{\rho} \hat{P}_e,
\] (1.66)

\[
\hat{\rho}_{gg} = \hat{P}_g \hat{\rho} \hat{P}_g,
\] (1.67)

\[
\hat{\rho}_{eg} = \hat{P}_e \hat{\rho} \hat{P}_g.
\] (1.68)
These operators correspond to the blocks of the matrix (1.56), singled out by the round brackets. Thus, the density matrix (1.56) can be written in a more compact form,

$$\dot{\rho} = \begin{bmatrix} \dot{\rho}_{ee} & \dot{\rho}_{eg} \\ \dot{\rho}_{ge} & \dot{\rho}_{gg} \end{bmatrix}.$$  \hspace{1cm} (1.69)

Equations for the corresponding sub-matrices of (1.69) can be obtained by acting with the appropriate projection operators on equation (1.61). We get

$$\dot{\rho}_{ee} = -\frac{i}{\hbar} \left( \hat{V}_{eg} \hat{\rho}_{ge} - \hat{\rho}_{eg} \hat{V}_{ge} \right) - \Gamma \hat{\rho}_{ee} - \gamma \left( \hat{\rho}_{ee} - \hat{\rho}_{ee}^{(0)} \right),$$  \hspace{1cm} (1.70)

$$\dot{\rho}_{gg} = -\frac{i}{\hbar} \left( \hat{V}_{ge} \hat{\rho}_{eg} - \hat{\rho}_{ge} \hat{V}_{eg} \right) - \Gamma \sum_{q=-1,0,1} \hat{Q}_{ge}^{(q)} \hat{\rho} \hat{Q}_{eg}^{(q)} - \gamma \left( \hat{\rho}_{gg} - \hat{\rho}_{gg}^{(0)} \right),$$  \hspace{1cm} (1.71)

$$\dot{\rho}_{eg} = -\frac{i}{\hbar} \left( \left[ \hat{H}_0, \hat{\rho}_{eg} \right] + \hat{V}_{eg} \hat{\rho}_{gg} - \hat{\rho}_{ee} \hat{V}_{eg} \right) - \frac{\Gamma}{2} \hat{\rho}_{eg} - \gamma \left( \hat{\rho}_{eg} - \hat{\rho}_{eg}^{(0)} \right).$$  \hspace{1cm} (1.72)

When deriving equations (1.70–1.72) we used the fact that projection operators are idempotent, namely $\hat{P}_e \hat{P}_e = \hat{P}_e$, $\hat{P}_g \hat{P}_g = \hat{P}_g$, and that $\hat{P}_e + \hat{P}_g = \hat{1}$, where $\hat{1}$ is the identity matrix. The convention of indexing operators in (1.70–1.72) is the same as in (1.66–1.67), for example $\hat{V}_{eg} = \hat{P}_e \hat{V} \hat{P}_g$.

Now, we will find stationary solution describing asymptotic behavior of the DTLS subject to a strong field of the pump beam. We take the potential in the form

$$\hat{V} = \hat{V}^{(0)} = -E_0 e^{-i\omega t} \hat{e}_0 \cdot \hat{d} + \text{c.c.} = \hat{V}_{eg} e^{-i\omega t} + \hat{V}_{ge} e^{i\omega t},$$  \hspace{1cm} (1.73)

and write the density matrix elements in a rotating frame of reference:

$$\dot{\rho}_{ee} = \dot{\sigma}_{ee} \quad \dot{\rho}_{eg} = \dot{\sigma}_{eg} e^{-i\omega t} \quad \dot{\rho}_{ge} = \dot{\sigma}_{ge} e^{i\omega t}.$$  \hspace{1cm} (1.74)

In the above equations, $\dot{\sigma}$ is the slowly varying envelope. We also assume that in equilibrium ($t \to \infty$) time derivatives $\dot{\sigma}$ vanish. Introducing (1.73) and (1.74–1.75) to equations (1.66–1.67), which amounts to treating the master equation in a rotating-wave approximation [25, 29], we obtain a set of algebraic equations for the individual matrix elements of $\sigma$:

$$(\Gamma + \gamma) \sigma_{m_e m'_e} = -\frac{i}{\hbar} \sum_{m_g = -F_g}^{F_g} \left( \mathcal{V}_{m_e m_g} \sigma_{m_g m'_e} - \sigma_{m_e m_g} \mathcal{V}_{m_g m'_e} \right) + \gamma \rho_{m_e m'_e}^{(0)},$$  \hspace{1cm} (1.76)

$$\gamma \sigma_{m_g m'_g} = -\frac{i}{\hbar} \sum_{m_e = -F_e}^{F_e} \left( \mathcal{V}_{m_g m_e} \sigma_{m_e m_g} - \sigma_{m_g m_e} \mathcal{V}_{m_e m_g} \right) +$$

$$+ \Gamma \sum_{q=-1,0,1} \mathcal{C}_{F_g m_g m'_g}^{F_g m_g m'_g} \sigma_{m_e = m_g + q, m'_e = m'_g + q} + \gamma \rho_{m_g m'_g}^{(0)},$$  \hspace{1cm} (1.77)

$$\left( \frac{\Gamma}{2} + \gamma - i\Delta \right) \sigma_{m_e m_g} = -\frac{i}{\hbar} \sum_{m'_e = -F_e}^{F_e} \mathcal{V}_{m_e m'_e} \sigma_{m'_e m_g} + \frac{i}{\hbar} \sum_{m'_e = -F_e}^{F_e} \sigma_{m_e m'_e} \mathcal{V}_{m'_e m_g}$$

$$+ \gamma \rho_{m_e m_g}^{(0)},$$  \hspace{1cm} (1.78)
In equation (1.77), $C^{F_{m_g m_{g'}}}_{F_{m_g m_{g'}}}$ is the product of the Clebsh-Gordan coefficients connected with corresponding energy levels. Solution of the equations (1.76–1.77) describe the stationary response of the DTLS perturbed by the strong pump field. Note, that except for the rotating-wave approximation, we have not made any further approximations, hence this solution is exact. We shall find the response of the pump-perturbed DTLS to the weak probe field, taking into consideration only the dominant Fourier amplitudes.

The probe beam absorption is governed by terms oscillating at the probe beam frequency $\omega_1$, namely $\hat{\sigma}_{eg} = \hat{\sigma}_{eg}^\dagger$, and the four-wave mixing signal originates from terms $\hat{\sigma}_{eg} = \hat{\sigma}_{ge}^\dagger$ evolving at frequency $2\omega - \omega_1$. By introducing the solution forms given by (1.80-1.81) to (1.70-1.71), keeping in mind that all time derivatives of $\hat{\sigma}$ vanish and dropping the terms which oscillate at frequencies different from frequency of a corresponding $\hat{\sigma}$ matrix element, we arrive at the following equations\textsuperscript{16}

\[
\hat{V} = \hat{V}^{(0)} + \hat{V}^{(1)} = \hat{V}_{eg} e^{-i\omega t} + \hat{V}_{ge} e^{i\omega t} + \hat{V}_{eg}^{(1)} e^{-i\omega t} + \hat{V}_{ge}^{(1)} e^{i\omega t},
\]

and write solutions in the general form

\[
\hat{\rho}_{ee} = \hat{\sigma}_{ee} + \hat{\sigma}_{eg}^\dagger e^{i\delta t} + \hat{\sigma}_{ee} e^{-i\delta t}, \quad \hat{\rho}_{eg} = (\hat{\sigma}_{eg} + \hat{\sigma}_{eg}^\dagger e^{i\delta t} + \hat{\sigma}_{eg} e^{-i\delta t}) e^{-i\omega t},
\]

\[
\hat{\rho}_{gg} = \hat{\sigma}_{gg} + \hat{\sigma}_{ge} e^{i\delta t} + \hat{\sigma}_{gg} e^{-i\delta t}, \quad \hat{\rho}_{ge} = (\hat{\sigma}_{ge} + \hat{\sigma}_{ge}^\dagger e^{i\delta t} + \hat{\sigma}_{ge} e^{-i\delta t}) e^{i\omega t}.
\]

Such a form implies the 1st order treatment of the probe field, similar to that used in Section 1.12. Indeed, the steady-state oscillations of optical coherences that are present in equations (1.80–1.81) occur at three basic frequencies: pump frequency $\omega$, probe frequency $\omega + \delta = \omega_1$ and four-wave mixing frequency $\omega - \delta = 2\omega - \omega_1$.

Taking into consideration probe field to all orders would result in infinite number of oscillation frequencies, namely $n\omega \pm m\omega_1$, where $n, m$ are integers [40].

One can also interpret equations (1.80-1.81) as Fourier expansion of the time-dependent density matrix $\hat{\rho}$, with elements of $\hat{\sigma}$ matrix being Fourier amplitudes of relevant oscillations. The first-order probe treatment is equivalent to taking into consideration only the dominant Fourier amplitudes.

The probe beam absorption is governed by terms oscillating at the probe beam frequency $\omega_1$, namely $\hat{\sigma}_{eg} = \hat{\sigma}_{eg}^\dagger$, and the four-wave mixing signal originates from terms $\hat{\sigma}_{eg} = \hat{\sigma}_{ge}^\dagger$ evolving at frequency $2\omega - \omega_1$. By introducing the solution forms given by (1.80-1.81) to (1.70-1.71), keeping in mind that all time derivatives of $\hat{\sigma}$ vanish and dropping the terms which oscillate at frequencies different from frequency of a corresponding $\hat{\sigma}$ matrix element, we arrive at the following equations\textsuperscript{16}

\[
(\Gamma + \gamma + i\delta) \sigma_{m_e m'_e}^{\dagger} = -\frac{i}{\hbar} \sum_{m_g = -F_g}^{F_g} (C_{m_e m_g}^{m_e m'_{e}} \sigma_{m_e m'_e}^{\dagger} - \sigma_{m_e m_g}^{\dagger} V_{m_g m'_e}) - \sigma_{m_e m_g}^{\dagger} V_{m_e m'_g}^{(1)}
\]

\[
\left(\frac{\Gamma}{2} + \gamma + i(\delta - \Delta)\right) \sigma_{m_e m_g}^{\dagger} = -\frac{i}{\hbar} \left( \sum_{m'_g = -F_g}^{F_g} V_{m_e m_g}^{m'_g m'_e} \sigma_{m_e m'_g}^{\dagger} - \sum_{m'_e = -F_e}^{F_e} \sigma_{m_e m'_g}^{\dagger} V_{m'_e m_g}^{m'_e m_g} \right),
\]

\textsuperscript{16}Note that we have already solved the master equation for $\hat{\sigma}_{ee}$, $\hat{\sigma}_{eg}$ and $\hat{\sigma}_{gg}$. The elements of these matrices appear in equations (1.82-1.85) as the known quantities. The only unknown variables are the terms indexed with “$+$” and “$-$”.

\textsuperscript{16}
(\frac{\Gamma}{2} + \gamma + i(\delta + \Delta)) \sigma^+_{m_g m_e} = -\frac{i}{\hbar} \left( \sum_{m'_e = -F_e}^{F_e} \left( \mathcal{V}_{m_g m'_e} \sigma^+_{m'_e m_e} + \mathcal{V}^{(1)}_{m_g m'_e} \sigma_{m_e m'_e} \right) + \sum_{m'_g = -F_g}^{F_g} \left( \sigma^+_{m'_g m_e} \mathcal{V}_{m'_g m'_e} + \sigma_{m_g m'_e} \mathcal{V}^{(1)}_{m'_g m'_e} \right) \right), \quad (1.84)

(\gamma + i\delta) \sigma^+_{m_g m'_g} = -\frac{i}{\hbar} \sum_{m_e = -F_e}^{F_e} \left( \mathcal{V}^{(1)}_{m_g m_e} \sigma_{m_e m'_e} + \mathcal{V}_{m_g m'_e} \sigma^+_{m_e m'_e} - \sigma^+_{m_g m'_e} \mathcal{V}_{m_e m'_e} \right) + \Gamma \sum_{q = -1, 0, 1} \mathcal{C}_{F_g m_g m'_g q} \sigma^+_{m_e m'_e}, \quad (1.85)

1.12 Calculation of the absorption and four-wave mixing signals for DTLS

The set of algebraic equations (1.82–1.85) for elements of \( \hat{\sigma} \) (the envelope of \( \hat{\rho} \)) contains complete information about response of DTLS to the pump-probe laser field. Since absorption of the probe is governed by imaginary part of the susceptibility oscillating at the probe beam frequency \( \omega_1 = \omega + \delta \), we can calculate the probe absorption signal according to the formula

\[
I_{\text{ABS}} \sim \text{Im} \sum_{q = -1}^1 \text{Tr} \left( \hat{\sigma}^+_{ge} \hat{d}_{eg}^q \right). \quad (1.86)
\]

The susceptibility that oscillates at frequency \( 2\omega - \omega_1 = \omega - \delta \) is responsible for generation of the four-wave mixing signal. We measure the intensity of light created in this nonlinear process, which is calculated as

\[
I_{\text{FWM}} \sim \sum_{q = -1}^1 \left| \text{Tr} \left( \hat{\sigma}^+_{eg} \hat{d}_{ge}^q \right) \right|^2. \quad (1.87)
\]

The rubidium transition examined in this thesis occurs between states with \( F_g = 3 \) and \( F_e = 4 \), hence we have to cope with 256-element matrices which are almost impossible to handle without a computer. Therefore, the problem is treated numerically by the program written by Aurél Gábris\footnote{E-mail: gabrisa@optics.szfk.i.kfki.hu, WWW site url: http://bird.szfki.kfki.hu/~gabrisa/_index.html.} [44]. Unless otherwise stated, the pump-probe spectra presented further in this chapter are outputs of this program.

The DTLS approach to the calculation of atomic spectra can also be found in the series of Bo Gao papers [45]-[47] and in Lezama et al. [42, 48].

1.13 Two-level effects in DTLS

Some pump-probe polarization configurations allow to study the DTLS as the two-level system or, at least, the set of two-level systems.

Let us consider the \( F_g = 3 \rightarrow F_e = 4 \) transition and situation when polarization of the pump beam is circular (\( \sigma^+ \)) with the quantization axis chosen to be parallel...
to the pump direction of propagation. Thus, the atom is being optically pumped to the $|F_g = 3, m_e = 3\rangle$ sublevel of the ground manifold. If we probe such a perturbed DTLS with the probe beam of the same polarization $\sigma^+$ as the pump, we induce transitions between the $|F_g = 3, m_e = 3\rangle$ and $|F_e = 4, m_e = 4\rangle$ levels of the DTLS (see inset in Figure 1.15). It is now evident that the $\sigma^+-\sigma^+$ pump-probe configuration allows one to treat the DTLS in terms of the single two-level system.

Figure 1.15: The probe-beam absorption spectrum generated for the $F_g = 3 - F_e = 4$ DTLS perturbed by the $\sigma^+$-polarized pump and probed by $\sigma^+$-polarized probe. The inset shows that optical pumping to the $|F_g = 3, m_g = 3\rangle$ sublevel allows treatment of the DTLS as a two-level system. Transitions induced by the pump and probe are represented by thick and thin arrows, respectively. The corresponding spectrum was generated for the detuning $\Delta = -3\Gamma$, Rabi frequency $\Omega = 6\Gamma$ and $\gamma = 10^{-4}\Gamma$. One can verify that positions of the absorption and gain sidebands coincide with the general Rabi frequency $\pm \Omega' = \pm \sqrt{6^2 + 3^2}\Gamma \approx \pm 6.7\Gamma$.

Before going further, let us recall definition of the Rabi frequency associated with transition between the ground ($F_g$) and excited ($F_e$) level of DTLS with photon of polarization $q$. It is given by the following formula [43]

$$\Omega_q = \frac{-2E_q(\langle F_g||\hat{d}||F_e\rangle}{\hbar},$$

(1.88)

where $E_q$ is the $q$-th component of the electric field of incident pump in the spherical basis. For the DTLS, we can also express the Rabi frequency between two Zeeman sublevels $|F_g, m_g\rangle$ and $|F_e, m_e\rangle$, namely [43]

$$\Omega_{m_e m_g} = (-1)^{m_e-m_g}(\langle F_e, m_e, F_g, 1, m_g, m_e - m_g\rangle|\Omega_{-(m_e-m_g)}$$

(1.89)

with $\Omega_{-(m_e-m_g)}$ given by (1.88). In other words, the Rabi frequency between specific Zeeman sublevels is a fraction of Rabi frequency $\Omega_q$ (1.88) proportional to the relevant Clebsch-Gordan coefficient.

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18For example, if the pump beam is $\sigma^+$ polarized, $E_{-1} = E_0 = 0, E_1 = E$. 

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Now, let us return to the case of DTLS in the $\sigma^+ - \sigma^+$ pump-probe configuration. The Clebsch-Gordan coefficient associated with the pair of extreme Zeeman sublevels is always 1 (for integer $F$). Thus, determination of sideband positions in the absorption spectra for a given detuning from atomic resonance allows determination of the Rabi frequency (1.88)\(^{19}\). As an example, absorption spectrum calculated for the $F_g = 3 \leftrightarrow F_e = 4$ transition is depicted in Figure 1.15. As it can be seen, the shape of this spectrum fully agrees with the one calculated for the two-level system (Section 1.4, Figure 1.7). Also, the four-wave mixing signal for the $\sigma^+ - \sigma^+$ polarization configuration is the same as for two-level system (Figure 1.9, page 17).

Another configuration for pump-probe spectroscopy, which can be treated in terms of the two-level atom, is when both pump and probe polarizations are linear and parallel to quantization axis ($\pi$). In this situation, however, we do not consider the single two-level transition, as we did in the preceding discussion, but the set of two-level atoms, each of them with the Rabi frequency proportional to the relevant Clebsch-Gordan coefficient. In order to predict the shape of the probe absorption spectrum one has to calculate the population distribution of DTLS due to optical pumping, generate the two-level pump-probe spectra for the corresponding Rabi frequencies and add them weighted by population differences between $|F_e, m_e\rangle$ and $|F_g, m_g\rangle$. Populations and Rabi frequencies between pairs of Zeeman sublevels coupled by $\pi$ polarization in the $F_g = 3 \leftrightarrow F_e = 4$ DTLS are depicted in Figure 1.16. The two-level probe absorption spectra were obtained using the formulas from Ref. [40] and then added with appropriate weights. The result was then compared with the spectrum generated by the program describing the full system [44] and showed perfect agreement. Figure 1.17a depicts all two-level contributions (already multiplied by weighting factors) and the net absorption spectrum for the discussed

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\(^{19}\)Provided ground state relaxation is low enough to allow for the efficient optical pumping.
Figure 1.17: The probe-beam absorption (a) and four-wave mixing spectrum (b) generated for the $F_g = 3 - F_e = 4$ DTLS perturbed by $\pi$-polarized pump beam and probed by the probe beam of the same polarization. The detuning of the pump beam is $\Delta = -3\Gamma$, Rabi frequency $\Omega = 6\Gamma$ and $\gamma = 10^{-4}\Gamma$. The weighted summation of the two-level spectra results in the net spectra which fully overlap with the spectra generated with program [44].
1.14. Multilevel effects in the DTLS

In the previous section we considered the pump-probe polarization configuration which allows treatment of the DTLS either as a pure two-level system or as a set of various two-level systems. Now we shall concentrate on several pump-probe polarization configurations that reveal interesting physics associated with the multilevel nature of the DTLS.

First, let us recall the results discussed in Section 1.3. The dressed levels of the two level system within one manifold $|\pm\rangle_N$ are separated by generalized Rabi frequency $\Omega' = \sqrt{\Omega^2 + \Delta^2}$. In the limiting case of a weak interaction ($\Omega \ll |\Delta|, \Gamma$) this separation equals

$$\Omega' \approx \Delta \left( 1 + \frac{1}{2} \frac{\Omega^2}{\Delta^2} \right),$$

(1.90)

whereas in the case when the interaction is strong, namely for $\Omega \gg |\Delta|, \Gamma$, we have

$$\Omega' \approx |\Omega|.$$

(1.91)

Now, let us consider the DTLS perturbed by the pump beam. When the pump beam is linearly polarized ($\pi$), photons of the pump beam couple pairs of $|F_g, m_g\rangle$ and $|F_e, m_g\rangle$ sublevels. In the case of $\sigma^\pm$ polarization, pairs of $|F_g, m_g\rangle$ and $|F_e, m_g \pm 1\rangle$ are coupled. For each pair of levels, Rabi frequencies (1.89), and hence separations (1.90) or (1.91) are different. We have already taken this into account when deriving signals depicted in Figure 1.17, now we would like to focus on a different aspect of this fact.

Since each pair of levels is characterized by the different generalized Rabi frequency, the light shifts of adjacent Zeeman sublevels are different. For example, in the strong interaction limit and for $\pi$-polarized pump beam we have

$$\Delta_{|F_g, m_g\rangle, |F_e, m_g\rangle} = \frac{1}{2} ||\Omega_{m_e m_g}|| - ||\Omega_{m_e+1 m_g+1}|| = \frac{1}{2} ||\Omega|| |\mathcal{C}_m^{m_g} - \mathcal{C}_m^{m_g+1}|,$$

(1.92)

where $k$ is an integer and $\mathcal{C}_m^{m'}$ is the Clebsch-Gordan coefficient between the $|F_g, m\rangle$ and $|F_e, m'\rangle$ levels. Hence, the DTLS degeneracy is removed due to light shifts of the Zeeman sublevels. This fact affects the absorption and four wave mixing spectra.
Figure 1.18: The light-shifted sublevels of the DTLS perturbed by a strong $(\Omega \gg |\Delta|, |\Gamma|)$ $\pi$-polarized (a) and $\sigma^+$-polarized (b) pump beam. Thin dotted lines mark positions of unperturbed DTLS sublevels, thick solid lines are the DTLS sublevels shifted by interaction with the pump beam. Thin and thick bars are separated by Rabi frequencies $\Omega'_{m_g,m_e}$, Block arrows show directions of the light shifts (for $\Delta < 0$), thick grey arrows represent pump photons of appropriate polarization. The energy scale is not conserved (this is marked by the brake sign on the arrows), except for the relative light shifts which correspond to the real situation.
registered for appropriate pump-probe polarization configurations. The structure of the DTLS with the light-shifted sublevels for \( \pi \)- and \( \sigma^+ \)-polarized strong pump beam (\( \Omega \gg |\Delta|, \Gamma \)) is depicted in Figure 1.18.

### 1.14.1 \( \sigma^+ - \pi \) pump-probe configuration

Let us now consider the case when the pump and probe beams are \( \sigma^+ \) and \( \pi \) polarized, respectively. The pump beam prepares DTLS as shown in Figure 1.18b with population optically pumped to the \( |F_g, m_g = 3\rangle \) sublevel. The probe absorption spectrum computed for this configuration is depicted in Figure 1.19. The most characteristic feature of this spectrum is the lack of the gain sideband on the red side of the pump beam frequency and a deep, narrow absorption resonance in the vicinity of the pump beam frequency.

![Figure 1.19: The probe-beam absorption spectrum generated for the \( \sigma^+ - \pi \) pump-probe polarization configuration (\( \Omega = 6\Gamma \), \( \Delta = -3\Gamma \) and \( \gamma = 10^{-4}\Gamma \)).](image)

The appearance of the Rabi absorptive sideband and lack of the gain sideband can be easily understood in terms of the dressed atom model, keeping in mind that pump and probe photons can induce transitions with \( \Delta m = m_e - m_g = 1 \) and \( \Delta m = 0 \), respectively. Let us look at Figure 1.20. Absorption of the probe beam, which is one-photon process [25], starts from the \( |F_g, m_g = 3\rangle \) level and excites the atom to \( |F_e, m_e = 3\rangle \) (thicker vertical arrow in Figure 1.20). Since both levels are light-shifted, absorption occurs at the probe-pump detuning \( \delta = \frac{1}{2}(\Omega'_{m_e=3,m_g=2} + \Omega'_{m_e=4,m_g=3}) \). Gain for the probe beam is possible only for the transition between the \( |F_g, m_g = 3\rangle \) and \( |F_e, m_e = 3\rangle \) sublevels (thicker grey slant arrow). Recalling that gain is a three-photon process, in which two pump photons are absorbed and one probe photon is emitted [25] and remembering that pump photons are \( \sigma^+ \) polarized (thicker grey dashed arrows), we can easily understand that this process is impossible: there is no population in the \( |F_e, m_e = 2\rangle \) sublevel. However, a small amount of population occupies the dressed states \( |F_e, m_g = 4\rangle \). Thus, if we reverse the directions of the grey arrows in the Figure 1.20, we will see that another three photon process is possible - emission of two \( \sigma^+ \) pump photons and absorption of one \( \pi \) probe photon. This results in the faint absorption feature which occurs for the probe-pump detuning \( \delta = -\frac{1}{2}(\Omega'_{m_e=3,m_g=2} + \Omega'_{m_e=4,m_g=3}) \).
A dispersion-like, small resonance at the frequency of the pump beam is due to the Rayleigh resonance \[25\] between two \( |F_g, m_g = 3 \rangle \) and two \( |F_e, m_e = 4 \rangle \) sublevels\(^{20}\) belonging to different manifolds \( N \) and \( N + 1 \) of the dressed atom.

To understand the origin of the narrow resonance on the blue side of the pump beam frequency we shall employ the idea of Raman transitions \[25, 37\] depicted schematically in Figure 1.21. Two atomic levels, \( |1 \rangle \) and \( |2 \rangle \) (via level \( |v \rangle \), which can be virtual), separated by energy \( \hbar \delta_{21} \), induced by two laser beams of frequencies \( \omega_1 \) and \( \omega_2 \), occurs when the resonance condition \( \omega_2 - \omega_1 = \delta_{21} \) is fulfilled. The direction and the amplitude of the process are governed by the population difference. The process starting from more populated level (black arrows) predominates the reverse one (grey arrows).

For the sake of transparency the latter process is not shown in Figure 1.20.
fields are chosen so that $\omega_2 - \omega_1 = \delta_{21}$, the transition between $|1\rangle$ and $|2\rangle$ via level $|v\rangle$ becomes possible and the resonance occurs with the absorption of a photon from one beam and emission of a photon to the other beam. The direction of the process (from $|1\rangle$ to $|2\rangle$ and vice versa) is governed by the initial population of the levels - the process starting from the more populated level outweihts the reverse one, hence its amplitude is proportional to the population difference of states $|1\rangle$ and $|2\rangle$. The discussed process occurs even if $|v\rangle$ is a virtual state.

![Diagram](image)

**Figure 1.22:** Central part of the pump-probe spectra for the $\sigma^+ - \pi$ (like in Figure 1.19) and $\sigma^+ - \sigma^-$ pump-probe beams polarization configuration. The spectra were generated for $\Omega = 6\Gamma$, $\Delta = -3\Gamma$ and $\gamma = 10^{-4}\Gamma$. The absorption resonance for $\sigma^-$-polarized probe is two times farther from the $\delta = 0$ position than the resonance for the $\pi$-polarized probe. The inset explains this feature by the Raman transitions between the light-shifted sublevels of the ground level of the DTLS. Note that both resonances have subnatural, i.e. smaller than $\Gamma$, width.

Now, let us consider the light-shifted sublevels of the $F_g$ level of our DTLS, shown in Figure 1.20. Having pump and tunable probe beams at our disposal, we can perform Raman transitions between these sublevels. Since the only populated sublevel is $|F_g, m_g = 3\rangle$, the pump-probe polarizations allow only the transition from $|F_g, m_g = 3\rangle$ to $|F_g, m_g = 2\rangle$ with the absorption of a photon from the probe beam and emission of a photon to the pump beam. As discussed in the above paragraph and according to the considerations on the page 1.14 [equation (1.92)], the resonance condition reads

$$\delta = \frac{1}{2} |\Omega'_{m_e=4, m_g=3} - \Omega'_{m_e=3, m_g=2}|.$$  \hspace{1cm} (1.93)

Usually, $\delta$ calculated from (1.93) is a fraction of $\Gamma$. Since the Raman transition occurs between the dressed sublevels that mainly originate from the ground-state sublevels, the widths of the corresponding resonances are subnatural. These two facts explain the features of the narrow central absorption resonance in Figure 1.19. Figure 1.22
depicts the expanded central part of the spectrum 1.19 for the $\sigma^+ - \pi$ and of the spectrum calculated for the $\sigma^+ - \sigma^-$ pump-probe polarizations. In the latter case, the Raman process ends at the $|F_g, m_g = 1\rangle$ sublevel, which is shifted with respect to the $|F_g, m_g = 3\rangle$ sublevel approximately twice as much as the $|F_g, m_g = 2\rangle$ sublevel. Thus, the position of the corresponding resonance is approximately two times farther from the center of the spectra ($\delta = 0$) than the resonance connected with the $|F_g, m_g = 3\rangle$ to $|F_g, m_g = 2\rangle$ Raman transition. The inset in Figure 1.22 shows the scheme of both Raman transitions.

Finally, let us note that for the pump-probe beam polarization configurations discussed in this subsection the four-wave mixing process does not occur. One can easily understand this by trying to draw the four-wave mixing linkage of Figure 1.10 between sublevels of the considered DTLS. This cannot be done without violation of the transition rules.

The theoretical predictions presented in this subsection were experimentally verified in Refs. [49, 50, 51] and interpretation of the subnatural resonant structures observed around the pump beam frequency in terms of Raman transitions between light-shifted sublevels of the DTLS was first given in [49].

1.14.2 $\pi - \sigma^+$ pump-probe configuration

Let us consider an atom subject to a $\pi$-polarized pump beam and probed by a $\sigma^+$-polarized beam. The $\pi$ light shifts the atomic sublevels and creates alignment, i.e. symmetric distribution of populations with respect to the $|m_g = 0\rangle$ sublevel. We shall focus on the ground state sublevels depicted in Figure 1.23.

In contrast to the case discussed in the previous section, here all ground-state sublevels are populated and give rise to the spectra. Depending on the sign of probe-pump detuning $\delta$, two Raman processes involving pump and probe photons are possible: gain of the probe, which happens for $\delta < 0$ and absorption of the probe for $\delta > 0$. The resonances occur every time $|\delta|$ coincides with the energy separation of the adjacent sublevels. The amplitude of the corresponding gain or the absorption resonances is proportional to the population difference of the sublevels involved in the transition and to a squared product of Clebsch-Gordan coefficients associated with the specific Raman transition. Such weighting causes the absorption feature of the Raman transition to always be smaller than the corresponding gain. For example, for the $|m_g = 0\rangle \leftrightarrow |m_g = 1\rangle$ transition (absorption), the amplitude is proportional to $(\frac{\sqrt{7}}{\sqrt{14}} \cdot \frac{\sqrt{15}}{\sqrt{14}})^2 \approx 0.19$, whereas for $|m_g = 0\rangle \leftrightarrow |m_g = -1\rangle$ (gain) we have $(\frac{2}{\sqrt{7}} \cdot \frac{3}{\sqrt{14}})^2 \approx 0.12$.

Now, we should be able to model the probe absorption spectrum by summing the Lorentzian profiles centered at appropriate resonance frequencies and weighted by products of the relevant population differences and squared Clebsch-Gordan coefficients, namely

$$s(\delta) = \sum_{i=1}^{3} w_{i-1,i} \Delta \Pi_{i-1,i} \mathcal{L}_{i-1,i}(\delta) - \sum_{i=-1}^{-3} w_{i+1,i} \Delta \Pi_{i+1,i} \mathcal{L}_{i+1,i}(\delta). \quad (1.94)$$

In equation (1.94) $w_{i-1,i}$ is the weight associated with the Clebsch-Gordan coefficients along the $|m_g = i - 1\rangle \leftrightarrow |m_g = i\rangle$ Raman transition path, $\Delta \Pi_{i-1,i}$ is the
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Figure 1.23: (a) Ground sublevels of the $F_g = 3 - F_e = 4$ DTLS perturbed by $\pi$-polarized beam (grey, thick arrow) and probed by $\sigma^+$-polarized beam (thin arrow). Due to population distribution, gain (absorption) of the probe beam occurs for $\delta < 0$ ($\delta > 0$). Relative light-shift of the sublevels was calculated for $\Omega = 6\Gamma$ and $\Delta = -3\Gamma$. (b) Resonances corresponding to Raman transitions between light-shifted DTLS sublevels plotted for $\gamma_{i-1,i} = 0.08\Gamma$, $\Omega = 6\Gamma$ and $\Delta = -3\Gamma$.

The signal calculated according to (1.94) was compared with the spectrum generated by the code of Ref. [44] (Figure 1.24). Despite qualitative similarity of the spectra, the differences connected with the widths of resonances in the spectra generated by [44] and (1.94) are quite pronounced. Since spectra (b) and (c) fit better to the experimental data of Section 4.4, page 82, we attribute this difference to the program inaccuracy.

Population difference of $|m_g = i - 1\rangle$ and $|m_g = i\rangle$, and

$$L_{i-1,i}(\delta) = \frac{\gamma_{i-1,i}}{\gamma_{i-1,i}^2 + (\delta - \delta_{i-1,i})^2}$$

where $\delta_{i-1,i}$ and $\gamma_{i-1,i}$ are, respectively, the resonance frequencies and widths of the $|m_g = i - 1\rangle \leftrightarrow |m_g = i\rangle$ transitions.
1.14.3 Configurations with multiple pump beams

In the previous section we have considered a single pump beam and a probe beam in a well-defined polarization configuration. Despite complexity of DTLS, we were able to predict, at least qualitatively, shapes of the probe absorption and four-wave mixing spectra. The situation becomes more complicated when atoms are perturbed by the net light field created by the interference of many pump beams of various polarizations and directions. This interference results in the spatial modulation of the light intensity and polarization which in turn gives rise to many interesting phenomena connected with atomic motion or localization. This problem will be discussed in more detail in the next section. Here we shall briefly discuss two common, one-dimensional configurations of the two counter-propagating pump beams: \( \sigma^+ - \sigma^- \) and \( \text{lin} \perp \text{lin} \) (Figure 1.25).

In the \( \sigma^+ - \sigma^- \) configuration amplitude of the net \( \mathbf{E} \) vector is constant, hence the light-shifts of atomic sublevels are space-independent. However, the direction of the electric field is helically modulated in space [52]. Thus, a travelling atom experiences rotation of the light polarization that leads to enhancement of atom cooling (the, so called, sub-Doppler cooling) [52]. The friction coefficient associated with this mechanism can be measured by means of pump-probe spectroscopy, as described in Ref. [53] and Section 4.4.7, page 99. The corresponding resonance appears as a narrow (width of the order of 100 kHz), dispersion-like structure in the center of Raman spectra associated with light-shifted sublevels of the DTLS.

The \( \text{lin} \perp \text{lin} \) configuration leads to a spatial modulation of polarization with period \( \lambda/2 \) (see Ref. [52] and Section 2.5, page 48). Hence, the light-shifts of Zeeman sublevels become space dependent. This gives rise to another mechanism of sub-Doppler cooling [52] and to atomic confinement in periodically distributed po-
Figure 1.25: Two one-dimensional pump beams configurations: (a) $\text{lin} \perp \text{lin}$ and (b) $\sigma^+ - \sigma^-$. The net field resulting from the beam interference interacts with the cold atomic medium which is then probed by the probe beam $k_{pr}$ of arbitrary polarization, usually making a small angle $\theta$ with one of the pump beams.

tential wells. Quantized motion of localized atoms leads to resonances that result from Raman transitions between discrete vibrational levels and appear in the probe absorption and four-wave mixing spectra [54].

Both for the $\sigma^+ - \sigma^-$ and $\text{lin} \perp \text{lin}$ configurations, resonances connected with the momentum exchange between atoms and the laser field occur. The, so-called, "recoil-induced resonances" are treated in detail in Chapter 2. A detailed theory of the probe transmission in the pump configurations discussed above can be found in [55, 56].

1.15 Conclusions

In this Chapter we discussed nonlinear effects that occur when an atom is perturbed by a strong, resonant laser field. We have shown how these effects are manifested in the probe and four-wave mixing spectra. First, we concentrated on a two-level atom, then we included effects of the level degeneracy. It was demonstrated that this degeneracy is responsible for narrow resonances occurring for small probe-pump detunings, $\delta \approx 0$, resulting from Raman transitions between the light-shifted atomic sublevels. In the last subsection of this Chapter, we mentioned that atomic motion influences the spectra in a very non-trivial way. If an atomic sample is sufficiently cold, the motion of atoms gives rise to new resonant structures which, although very narrow, can be resolved by high resolution Raman spectroscopy. This aspect will be further analyzed in Chapter 2.
Chapter 2

Influence of atomic motion on the spectra

2.1 Introduction

In Chapter 1 we considered transitions between internal, quantized energy levels of an atom. These transitions occur either between the ground and excited states of an atom or, in the case of Raman resonances, between light-shifted magnetic sublevels of the ground state. The former result in resonances with characteristic width of the order of the natural linewidth $\Gamma$, the latter appear in the spectra as ultra-narrow features with widths characterized by the decay rate of the ground level $\gamma$ and broadened by the excited states’ admixtures to the dressed states wave functions. In our previous considerations we neglected atomic motion, thus the results of Chapter 1 apply only to atoms at rest.

For standard atomic samples like vapor cells or atomic beams, atomic motion is generally a disturbing factor which broadens the spectra. In the case of laser cooled samples, however, a tremendous narrowing of the atomic velocity distribution not only suppresses the broadening, but also reveals interesting phenomena connected with the momentum transfer between atoms and laser beams. Moreover, light modulation in the region of beam overlap, due to interference, can lead to atomic confinement in potential wells spatially separated by a fraction of light wavelength. Such quantization is responsible for vibrational and oscillatory structures of the probe-beam spectra.

The momentum exchange between non-localized atoms and the laser field is associated with the phenomenon of the so-called recoil-induced resonances (RIRs). They were noticed for the first time in the spectra registered in experiment [54], but the first unambiguous observation of RIRs was performed in Ref. [57]. A simple theory of RIRs can be found in Refs. [54, 58, 59]. It explains the origin of the corresponding resonances in the probe absorption spectrum and gives an intuitive picture of the RIR process. This theory will be presented in the following sections in this chapter. The rigorous theory with inclusion of simple multilevel atomic structure which allows calculation of both the probe absorption and four-wave mixing signal due to RIR processes is presented in Refs. [60]-[64].

The periodic structures of atoms localized in the potential wells created by spatial light modulation are known as optical lattices. The first evidence of the periodically ordered atom structures bound by light and of the quantization of their motion was given in Refs. [54, 65]. Earlier, however, it was shown that cold atoms localize in the
field of three-dimensional standing wave to less than a fraction of a wavelength [66].

The proof for that was the observation of Dicke narrowing [67] of the spectral line emitted by localized atoms. The atomic localization in a standing wave is a direct consequence of cooling atoms below the Doppler limit [68]-[70] which was experimentally observed in Ref. [71] and theoretically explained in Ref. [52]. Quantization of the atomic motion leads to creation of the discrete kinetic vibrational levels. Raman transitions between these levels result in narrow resonances, whose widths are governed by the time an atom spends in a potential well before it undergoes tunnelling or is removed from the well by collisions with non-localized particles.

Both transitions due to momentum exchange between non-localized atoms (RIR) and between the vibrational levels of optical lattices modify the spectra in a very close vicinity of the trapping (pump) laser frequency. In a general case, all these contributions act simultaneously, and one is confronted with a very complicated spectra with superpositions of many, sometimes overlapping, resonances around $\delta = 0$.

2.2 Principle of the recoil-induced resonances

Let us consider a situation such as depicted in Figure 2.1. A weak probe beam (with wave vector $k_{pr}$ and frequency $\omega + \delta$) makes an angle $\theta$ with the direction of propagation of the strong pump beam (wave vector $k$, frequency $\omega$). The two beams overlap in the cold-atom sample. We consider absorption of photons from one of the beams followed by stimulated emission to the other beam. Such a process results in the momentum change $\Delta p$ of an atom.

![Figure 2.1: Geometry for observation of the recoil-induced resonances.](image)

We assume that atoms can move freely, at least along direction of the momentum exchange $\hat{y}||\Delta p$, so there is no potential that localizes atoms in this direction. The probe-pump detuning is very small, hence with a good accuracy we can write $|k| = |k_{pr}| \approx k$. 
In the process of pump photon absorption followed by the probe photon emission, the change of atomic momentum is given by

\[ \Delta p = p_{\text{final}} - p_{\text{initial}} = p_{\text{initial}} + \hbar(k - k_{pr}) - p_{\text{initial}} = -2\hbar k \sin \frac{\theta}{2} \hat{y}. \]  

(2.1)

For a given angle \( \theta \) the momentum transfer processes do not change components of the atomic momentum other than \( p_y = p \cdot \hat{y} \), hence, we can treat our problem in one dimension. Thereby, we can rewrite equation (2.1) in a scalar form

\[ \Delta p = -2\hbar k \sin \frac{\theta}{2}. \]  

(2.2)

The corresponding change of atomic kinetic energy is equal

\[ \Delta E_{\text{kin}} = \frac{1}{2m} [(p_y + \Delta p)^2 - p_y^2] = \frac{2p_y \Delta p + \Delta p^2}{2m}, \]  

(2.3)

where \( m \) is the atomic mass. From equations (2.2,2.3) one can deduce that atoms which move initially with momentum

\[ p_y > -\Delta p/2 = \hbar k \sin \frac{\theta}{2} \]  

(2.4)

are decelerated (\( \Delta E_{\text{kin}} < 0 \)), while those with

\[ p_y < \hbar k \sin \frac{\theta}{2} \]  

(2.5)

experience acceleration (\( \Delta E_{\text{kin}} > 0 \)). Due to population difference between the continuum states of kinetic energy, governed by the Maxwell distribution, the process of acceleration dominates (Figure 2.2a). The resonance occurs every time the probe-pump detuning coincides with the kinetic energy difference (2.3), namely at detuning

\[ \delta_{\text{res}} = 2 \frac{k}{m} \sin \frac{\theta}{2} \left( \hbar k \sin \frac{\theta}{2} - p_y \right). \]  

(2.6)

Since in the case of a predominating acceleration process (\( p_y > \hbar k \sin \frac{\theta}{2} \)) \( \delta_{\text{res}} < 0 \), the gain of the probe beam is expected for negative probe-pump detuning.

Analogous consideration can be performed in the case of probe photon absorption followed by emission of a photon to the pump beam. In such a case, the process in which atoms with \( p_y > -\hbar k \sin \frac{\theta}{2} \) are accelerated predominates (Figure 2.2b). Hence we observe absorption of the probe for positive probe-pump detuning, \( \delta_{\text{res}} > 0 \).

Note, that we have assumed that we deal with two-level atoms, hence polarization of the pump and probe beams was not taken into account.

## 2.3 Calculation of the probe absorption signal

Conclusions from the previous section allow one to calculate the probe beam absorption signal due to momentum transfer processes. To start with, we shall model the resonance shape. We assume that the resonance is Lorentzian-like, with arbitrary chosen phenomenological width \( \gamma \), which reflects possible departures from the exact
Figure 2.2: Two mechanisms of RIRs due to (a) pump photon absorption followed by emission of a photon to the probe beam and (b) probe photon absorption followed by emission of a photon to the pump beam. Process (a) results in the probe gain, (b) leads to the probe absorption. Solid arrows represent processes which predominate over the reverse ones, marked by dotted arrows. The probe gain occurs for negative probe-pump detuning $\delta < 0$, absorption of the probe occurs for $\delta > 0$. Note, that the net effect, either for the probe gain or absorption result in acceleration of atoms, which is the consequence of the Maxwell velocity distribution.

conservation of the kinetic energy\(^1\). Hence the shape of the resonance can be written as

$$L(\delta, \delta_{\text{res}}, \gamma) = \frac{\gamma}{\gamma^2 + (\delta - \delta_{\text{res}})^2}, \quad (2.7)$$

where $\delta_{\text{res}}$ is given by (2.6). The resonance amplitude, as usual in the case of the Raman transitions (see Section 1.14.1, page 34), is proportional to the population difference between initial and final momentum states, namely

$$\Delta \Pi(p_y_{\text{final}}, p_y_{\text{initial}}) = \Pi(p_y + \Delta p) - \Pi(p_y), \quad (2.8)$$

where $\Pi(p)$ stands for the population of the continuum state with momentum $p$. In the case of thermal equilibrium we assume that $\Pi(p)$ is given by the Maxwell distribution

$$\Pi(p) = (2\pi k_B T)^{-\frac{3}{2}} \exp \left[ -\frac{p^2}{2mk_B T} \right], \quad (2.9)$$

where $T$ is the sample temperature and $k_B$ is the Boltzmann constant. In general, $\Pi(p)$ can be arbitrarily chosen.

In order to derive the absorption signal $s(\delta)$, we have to multiply the shape of resonance (2.7) by its amplitude (2.8) and average over the whole momentum range in our sample:

$$s(\delta) = -\int_{-\infty}^{\infty} dp_y \Delta \Pi(p_y + \Delta p, p_y) \ L(\delta, \delta_{\text{res}}, \gamma). \quad (2.10)$$

The integral in (2.10) can be carried out numerically, giving exact shape of the recoil-induced resonance with broadening parameter $\gamma$. For typical conditions in a

\(^1\)For example, $\gamma$ can be related to a finite interaction time of atoms with laser field [50].
standard magneto-optical trap, however, useful approximations allow one to derive analytic formula for $s(\delta)$, Ref. [58]. If the atoms are warm enough to fulfill the condition $\Delta p = 2\hbar k \sin \frac{\theta}{2} \ll p_T$, where

$$p_T = \sqrt{mk_B T}$$

is the momentum characterizing the width of thermal atomic distribution, we can approximate $\Delta \Pi$ by writing

$$\Delta \Pi (p_y + \Delta p, p_y) \approx \frac{\partial \Pi}{\partial p_y} \Delta p.$$ \hspace{1cm} (2.12)

For example, $\hbar k/p_T \approx 0.2$ for $T = 10 \mu K$ and $^{85}\text{Rb} D_2$ line, which means that the approximation holds even for such a low temperature. If $\gamma \ll k p_T / m \approx 2 \pi \cdot 40$ kHz (for the conditions), we can replace $\mathcal{L}(\delta, \delta_{\text{res}}, \gamma)$ in (2.13) by the Dirac delta function, which amounts to demanding exact conservation of kinetic energy:

$$\mathcal{L}(\delta, \delta_{\text{res}}, \gamma) \approx \delta_D (\delta - \delta_{\text{res}}).$$ \hspace{1cm} (2.13)

Introducing approximations (2.12,2.13) into (2.10), we obtain an analytic formula for the absorption signal which results from the momentum transfer process

$$s(\delta) = - \sqrt{\frac{m}{2\pi}} \frac{\hbar \delta}{2 u_T^3/2 k \sin(\theta/2)} \exp \left[ - \frac{\delta^2}{2 u_T^2(2k \sin(\theta/2))^2} \right],$$ \hspace{1cm} (2.14)

where $u_T = p_T / m$. The signal (2.14) is the derivative of a Gaussian function\(^2\) (Figure 2.3). It has a minimum for $\delta = 2k u_T \sin(\theta/2)$ and a maximum for $\delta = -2k u_T \sin(\theta/2)$. The width of the spectrum, defined as the distance between minimum and maximum, is proportional to a square root of $T$. The RIR spectrum can be thus regarded as a spectroscopic tool for measurement of the temperature of a cold atomic sample, according to the formula

$$T = \frac{m}{16k_B k^2 \sin^2(\theta/2) w^2},$$ \hspace{1cm} (2.15)

where $w$ is the measured signal width. Moreover, the method of the RIR spectroscopy offers a unique possibility of selective probing of the velocity distribution in the direction of the momentum exchange (in this case $\hat{y}$).

In many experiments it is convenient to have the pump and probe beams making small angle $\theta$. In this situation we can replace $\sin(\theta/2)$ in Ref. (2.14) by its argument and arrive at the formula derived in [58]. The drawback of such a setup is the reduced accuracy of the temperature determination relative to the case of a bigger angle. It is caused by high resolution requirements (the RIR signal becomes very narrow) and is highly sensitive to the angle determination ($T \propto \theta^{-2}$). In Ref. [50] this drawback has been eliminated by choosing $\theta = 90^\circ$.

### 2.4 Momentum exchange process in four-wave mixing

Derivation of the four-wave mixing signal due to recoil-induced resonances is not so straightforward as in the case of the probe absorption spectrum. The simplest

\(^2\)In general it could be derivative of an arbitrary velocity distribution function - see the remark below the formula (2.9).
Figure 2.3: Probe absorption signal due to recoil-induced resonances. The gain (absorption) of the probe beam occurs for negative (positive) detuning $\delta$, with the peak value for $\delta = -2ku_T \sin(\theta/2)$ ($\delta = 2ku_T \sin(\theta/2)$), $u_T$ being the most probable velocity of atoms in temperature $T$. Width of the signal is proportional to $\sqrt{T}$. The spectrum was generated for $T = 50 \mu K$ and $\theta = 10^\circ$. The signal broadening due to $\gamma$ is negligible, which was verified by comparison of numerical computation according to (2.10) with analytic result (2.14) for $\gamma$ up to 0.05 $\Gamma$.

situation of a two-level atom subject to two counter-propagating pump beams and a probe beam making angle $\theta$ with one of them (Figure 2.4) has been analyzed theoretically in Ref. [60].

To discuss momentum exchange processes, we have to consider density matrix which includes the center-of-mass momentum states, namely

$$\hat{\rho} = \sum_{\alpha,\beta=\{g,e\}} \rho_{\alpha\beta} (p, p') |\alpha, p\rangle \langle \beta, p'|,$$

(2.16)

where

$$\rho_{\alpha\alpha} = \tilde{\rho}_{\alpha\alpha} (p, p') e^{-i[(p^2-p'^2)/2\hbar m]t},$$

(2.17)

$$\rho_{ge} = \tilde{\rho}_{ge} (p, p') e^{-i[(p^2-p'^2)/2\hbar m]t} e^{i\omega t},$$

(2.18)
\( \omega \) being the frequency of the pump beams. We assume the ground state equilibrium distribution equal to

\[
\hat{\rho}^{(0)}_{gg}(p, p') = n(2\pi\hbar)^3 \frac{1}{(\sqrt{\pi}pt)^3} e^{-\mathbf{p}^2/2} \delta_D(p - p'),
\]

(2.19)

where \( n \) is the atomic density and \( pt \) – the most probable atomic momentum. Since we look for the macroscopic polarization of the medium given by formula (1.3), we are interested in the density matrix element \( \rho_{ge}(r, t) \) given by

\[
\rho_{ge}(r, t) = \frac{1}{(2\pi\hbar)^3} \int dp \int dp' e^{i\mathbf{p}' \cdot \mathbf{r}/\hbar} \rho_{ge}(p, p').
\]

(2.20)

According to Ref. [60], the third-order perturbation solution of the equation for the density matrix yields

\[
\rho^{(f)}_{eg}(r, t) = C \frac{P^{(f)}_{e(b)}}{\Gamma/2 + i\Delta} \left[ w\left( s^{(f)}_{(b)} \right) - \int dq N(q) w\left( z^{(f)}_{(b)} \right) \right] +
\]

\[
+ C \frac{P^{(f)}_{e(b)}}{\Gamma/2 - i(\Delta + \delta)} \left[ w\left( r^{(f)}_{(b)} \right) - \int dq N(q) w\left( z^{(f)}_{(b)} \right) \right] +
\]

(2.21)

where

\[
C = -iN_0 \frac{\Omega_1^* \Omega_2^* \Omega_{pr}}{\Gamma/2 + i(\Delta - \delta)} e^{i\mathbf{k}_m \cdot \mathbf{r} + i(\omega - \delta)}
\]

(2.22)

\[
P^{(f)}_{e(b)} = \begin{cases} 
(f) : \sqrt{\pi} \frac{2k\mu T \sin \theta/2}{2k\mu T \sin \theta/2} \\
(b) : \sqrt{\pi} \frac{2k\mu T \cos \theta/2}{2k\mu T \cos \theta/2} 
\end{cases}
\]

(2.23)

\[
s^{(f)}_{(b)} = \begin{cases} 
(f) : \frac{1}{2k\mu T \sin \theta/2} (i\gamma + \delta + 2\omega_k(1 - \cos \theta)) \\
(b) : \frac{1}{2k\mu T \cos \theta/2} (i\gamma + \delta + 2\omega_k(1 + \cos \theta))
\end{cases}
\]

(2.24)

\[
z^{(f)}_{(b)} = \begin{cases} 
(f) : \frac{1}{2k\mu T \sin \theta/2} \left( i\gamma + \delta + 2\omega_k \left[ \frac{q_x}{k} (1 - \cos \theta) - \frac{q_y}{k} \sin \theta \right] \right) \\
(b) : \frac{1}{2k\mu T \cos \theta/2} \left( i\gamma + \delta + 2\omega_k \left[ \frac{q_x}{k} (1 + \cos \theta) + \frac{q_y}{k} \sin \theta \right] \right)
\end{cases}
\]

(2.25)

\[
r^{(f)}_{(b)} = \begin{cases} 
(f) : \frac{1}{2k\mu T \sin \theta/2} (i\gamma + \delta - 2\omega_k(1 - \cos \theta)) \\
(b) : \frac{1}{2k\mu T \cos \theta/2} (i\gamma + \delta - 2\omega_k(1 + \cos \theta))
\end{cases}
\]

(2.26)

In equations (2.21-2.26) \( \Omega_i \) is the Rabi frequency associated with the \( i \)-th beam, \( k = |k_1| = |k_2| \cong |k_{pe}| \), \( \omega_k = \hbar k^2/2m \) is the so-called recoil frequency, \( q_i \) are the Cartesian components of the wave vector \( \mathbf{q} \) of a spontaneously emitted photon, \( N(\mathbf{q}) \) is the normalized probability density for spontaneous emission of photon having
momentum $\hbar q$ (in our calculations $N(q) = \frac{3}{8} \sin^2 \theta$), and $w(z)$ is the complex error function [72]

$$w(z) = \frac{i}{\pi} \int_{-\infty}^{\infty} e^{-t^2} z - t \, dt, \quad \text{Im}(z) > 0. \quad (2.27)$$

Indices $(f)$ and $(b)$ denote the “forward” and “backward” contributions, respectively. The “forward” contribution is due to scattering of photons of the $k_2$ beam on the population grating created by $k_1$ and $k_{pr}$ beams, whereas the “backward” contribution refers to the scattering of the $k_1$ beam on the $k_2$ and $k_{pr}$ grating. The four-wave mixing signal is then proportional to $|\rho_{ge}|^2$, where $\rho_{ge} = \rho_{ge}(f) + \rho_{ge}(b)$. The recoil-induced resonances in the four-wave mixing spectrum are shown in Figure 2.5. This spectrum has been calculated for the same parameters as the absorption spectrum depicted in Figure 2.3.

![Figure 2.5: Recoil-induced resonances in the four-wave mixing spectrum numerically generated according to formula (2.21) for $T = 50 \mu K$ and $\theta = 10^\circ$. Inset shows the squares of moduli of the individual “forward” and “backward” contributions. The “forward” contribution is responsible for the narrow feature in the spectrum.](image)

2.5 Localization of atoms – optical lattices

The notion of an optical lattice and atomic localization [73]-[76] is most easily understood when explained together with the sub-Doppler Sisyphus cooling mechanism [52]. Let us consider 1D example of an atom in the field of two counterpropagating laser beams of the same frequency $\omega$ and linearly polarized in orthogonal directions (Figure 2.7a). It is well known that for this so-called $\text{lin}\perp\text{lin}$ configuration the net electric field forms a standing wave

$$\mathbf{E}(z, t) = \frac{\sqrt{2}}{2} E_0 \mathbf{\hat{e}}(z) e^{-i\omega t} + c.c. \quad (2.28)$$

with space-modulated polarization unit vector $\mathbf{\hat{e}}(z)$

$$\mathbf{\hat{e}}(z) = \mathbf{\hat{e}}_X - i\mathbf{\hat{e}}_Y. \quad (2.29)$$
where $\hat{\epsilon}_X = (\hat{y} + \hat{x})/\sqrt{2}$ and $\hat{\epsilon}_Y = (\hat{y} - \hat{x})/\sqrt{2}$. It is straightforward to verify that for $z_+ = \frac{3}{8}\lambda + \frac{7}{8}\lambda$ the field is $\sigma^+$-polarized, for $z_- = \frac{1}{8}\lambda + \frac{5}{8}\lambda - \sigma^-$-polarized and for $z_{\text{lin}} = \frac{m}{4}\lambda - \pi$-polarized ($m$ is integer). Note, that the amplitude of the electric field remains space-independent and equal $2E_0$.

Now let us consider the $F_g = \frac{1}{2} \leftrightarrow F_e = \frac{3}{2}$ transition, depicted in Figure 2.6, subject to the field discussed above. Spatial modulation of the polarization is manifest in a space-dependent coupling of the atomic sublevels. For example, in the points where polarization of the net field is $\sigma^+$, the $| m_g = 1/2 \rangle$ level experiences energy shift three times bigger than the $| m_g = -1/2 \rangle$ level. Consequently, the potential energy of the $| m_g = -1/2 \rangle$ state in this point is higher than energy in the $| m_g = 1/2 \rangle$ state (Figure 2.7b).

![Figure 2.6: The $F_g = \frac{1}{2} \leftrightarrow F_e = \frac{3}{2}$ transition diagram with Clebsch-Gordan coefficients (in circles).](image)

According to Refs. [75, 55], the light-shift of a ground state sublevels resulting from such modulation, derived using the expansion of the generalized complex Rabi frequency for $\Omega \ll |\Delta|$, $\Gamma$ (see Appendix A), is given by the formula

$$U_{|\pm 1/2\rangle}(z) = \frac{U_0}{2} [2 \mp \sin(2kz)],$$

(2.30)

where $U_0$ is the Rabi frequency per beam, associated with the $| m_g = \pm 1/2 \rangle \leftrightarrow | m_g = \pm 3/2 \rangle$ transitions (i.e. those with Clebsch-Gordan coefficients equal to 1) and

$$U_0 = \frac{2}{3} \hbar \Delta \frac{\Omega_0^2/2}{\Delta^2 + \left(\Gamma/2\right)^2} = \frac{2}{3} \hbar \Delta s.$$  

(2.31)

In formula (2.31) we introduced saturation parameter $s$ (compare with the on-resonant saturation parameter (1.26) defined on page 9 and in Appendix A).

Let us focus our attention on an atom travelling in $+z$ direction and assume that at $z = \frac{1}{8}\lambda$ the atom is in state $| m_g = -1/2 \rangle$. For a given polarization configuration and negative detuning $\Delta < 0$, the atom has the lowest possible potential energy. In order to move further, the atom has to climb up a potential hill at the expense of its kinetic energy. In principle, the atom could regain its energy by falling down the potential hill from $z = \frac{3}{8}\lambda$ to $z = \frac{5}{8}\lambda$, but there is another possibility. If the atomic speed $v$ is slow enough, namely $v \simeq \frac{1}{4}\Gamma'\lambda$ where $\Gamma'$ is the optical pumping rate, the atom can be optically pumped to the $| m_g = 1/2 \rangle$ state before the next potential minimum, already in the vicinity of $z = \frac{3}{8}\lambda$. In this way the potential energy of the
atom is low again and its kinetic energy is decreased approximately by the height of the potential hill. The missing kinetic energy is dissipated in spontaneous emission of a $\pi$-polarized photon, blue-detuned with respect to $\omega$. This process can be repeated many times and results in a successive slowing down of the atoms. Finally, when the atomic kinetic energy is low enough, the atom is confined to one of the potential wells. Since the spatial distribution of the light-induced potential wells is periodic, we arrive at a periodically ordered ensemble of atoms, the so-called optical lattice. The period of such an optical lattice is determined by the light wavelength and beam geometry. In the one dimensional case discussed above, the period of the optical lattice equals $\frac{1}{4}\lambda$. The equilibrium temperature for atoms in an optical lattice can be roughly estimated by comparing the quantity $\frac{1}{2}k_B T$ with the depth of the optical potential given by (2.30), which yields

$$T_{\text{lattice}} \approx \frac{2}{3} \frac{\hbar \Omega_0^2}{|\Delta| k_B}. \quad (2.32)$$

Within an order of magnitude, the expression (2.32) agrees with the result of detailed calculations performed in Ref. [52]

$$T_{\text{lattice}} \approx \frac{1}{8} \frac{\hbar \Omega_0^2}{|\Delta| k_B}. \quad (2.33)$$

Equations (2.32, 2.33) show that by decreasing the light power and increasing its detuning we can lower temperature of atoms in the optical lattice below the Doppler
Localized atom as a quantum oscillator

Since the de Broglie wavelength of an atom trapped in the optical potential is comparable with the spatial size of the potential well, atomic motion becomes quantized and a structure of discrete vibrational energy levels appears. As atoms mostly occupy the bottom of potential surfaces, we can expand expression (2.30) around the minima $z_{\pm}$ and obtain

$$U_{|\pm 1/2\rangle}(z) \approx \frac{3}{2} U_0 - U_0 k^2 (z - z_{\pm})^2.$$  

(2.34)

The second term in (2.34) allows one to treat the atom in the potential well as a quantum harmonic oscillator, provided that the well is deep enough and atoms occupy the lowest-lying bound states. The equidistant vibrational energy levels of the oscillator are separated by the energy $\delta E_{\text{vib}} = \hbar \omega_{\text{vib}}$ where, according to (2.34)

$$\omega_{\text{vib}} = \frac{2}{\hbar} \sqrt{U_0 E_{\text{rec}}}.$$  

(2.35)

In (2.35) we introduced the recoil energy

$$E_{\text{rec}} = \frac{\hbar^2 k^2}{2m},$$  

(2.36)

which is the change of atomic kinetic energy after absorption or emission of a photon. Using quantum theory of the harmonic oscillator and Eq. (2.35), we can express the spatial extent $\delta z$ of the atomic wavefunction in the lowest bound vibrational state in terms of $E_{\text{rec}}$, namely

$$\delta z = \sqrt{\frac{\hbar}{2m \omega_{\text{vib}}}} = \frac{\lambda}{2\sqrt{2\pi}} \left( \frac{E_{\text{rec}}}{U_0} \right)^{1/4}.$$  

(2.37)

Spectroscopy of optical lattices

Quantization of atomic motion due to a tight confinement in optical potential can be detected by means of spectroscopy. Transitions between discrete vibrational levels were first observed in the probe transmission spectrum [54], in the fluorescence emitted by trapped atoms [65], and later in the four-wave mixing spectrum [77]. To detect transitions whose frequency is of the order of 100 kHz with optical fields, one has to employ Raman processes (page 34). In the probe transmission experiment two processes can occur: absorption of the a probe photon followed by emission of the lattice-field photon, or the reverse process (Figure 2.8a). The first process results in probe attenuation, the second leads to probe gain. If the probe-lattice field

3For comparison, let us recall that temperature associated with the Doppler cooling limit is given by $T_{\text{Dopp}} = h \Gamma / 2k_B$.

4For the $^{85}\text{Rb} D_2$ line, $E_{\text{rec}} = \hbar \omega_{\text{rec}}$, where $\omega_{\text{rec}} = 2\pi \cdot 3861 \text{ Hz}$.
detuning \( \delta = \omega_{pr} - \omega \) coincides with the vibrational frequency \( \omega_{vib} \) (or its multiple), a resonant Raman transition occurs. Since in thermal equilibrium vibrational levels are populated according to the Boltzmann distribution, the probe gain dominates for the negative detuning \( \delta < 0 \), while absorption of the probe is observed for \( \delta > 0 \). A typical probe absorption spectrum in an optical lattice is depicted in Figure 2.9.

\[
\delta = \omega_{pr} - \omega \quad \text{net probe absorption}
\]

\[
\delta = \omega_{pr} - \omega < 0 \quad \text{net probe gain}
\]

Figure 2.8: (a) Schematic representation of Raman processes involved in the probe transmission spectroscopy. Thick and thin arrows represent photons of the optical lattice field and probe beam, respectively. Transitions indicated by hollow arrows occur at lower rates than those indicated by black and grey arrows because of the Boltzmann population distribution of the vibrational levels (represented by dots of various sizes) (b) Schematic representation of a four-wave mixing on vibrational levels of optical lattice.

In the case of fluorescence spectroscopy of atoms in optical lattices, the lattice-field photons excite atoms from the initial vibrational state and spontaneous emission of photons brings the atoms back to the initial or to the neighboring vibrational level.\(^5\) In the first case, the scattered photons have frequency \( \omega \) of the lattice field, in the second, frequency \( \omega \pm m\omega_{vib} \) where \( m \) is an integer \((m \neq 0)\). Spectral analysis of the optical lattice fluorescence reveals a strong line due to elastic scattering at frequency \( \omega \) and sidebands at frequencies \( \omega \pm m\omega_{vib} \).

\(^5\)As it is shown experimentally in [65] and theoretically in [55], the elastic scattering of photons dominates (about 95% of all photons is scattered elastically).
Four-wave mixing processes result in generation of photons for the same resonance condition as in the case of probe absorption spectroscopy (Figure 2.8b). If the probe intensity is high enough, however, additional resonances at lower subharmonics of the vibrational frequency (at frequencies $\pm \frac{1}{2} \omega_{\text{vib}}, \pm \frac{1}{3} \omega_{\text{vib}}, \ldots$) can appear \cite{77}. This can be easily understood if we replace each single arrow representing the probe and lattice field photons in Figure 2.8b by their appropriate multiplicity.

Since resonance frequencies corresponding to vibrational transitions are very small compared to a spontaneous photon scattering rate $\Gamma' = s \Gamma$, it may seem impossible to resolve the corresponding resonances. However, it has been shown in Ref. \cite{55} that due to a tight confinement of an atom in optical potential, the $n$-th vibrational energy level decays at a rate

$$
\Gamma_n^{\text{eff}} \approx \Gamma' \sqrt{\frac{E_{\text{rec}}}{U_0}} \left( n + \frac{1}{2} \right) = \Gamma' \frac{2 E_{\text{rec}}}{\hbar \omega_{\text{vib}}} \left( n + \frac{1}{2} \right).
$$

(2.38)

Because the ratio of $E_{\text{rec}}$ to $\hbar \omega_{\text{vib}}$ is usually of the order of 30, the lifetime of low-lying bound states of the optical lattice is greatly enhanced so typical widths of vibrational resonances are about few tens of kHz to 100 kHz.

![Figure 2.9: Typical probe absorption spectrum in optical lattice with vibrational frequency $\pm \omega_{\text{vib}}$. Gain of the probe occurs for the negative detuning $\delta < 0$ and absorption for $\delta > 0$. The resonances are Lorentzian curves centered at $\pm \omega_{\text{vib}}$.](image)

### 2.8 Optical lattices in a MOT

In the previous sections we considered simple, one-dimensional configuration of orthogonally, linearly polarized, counter-propagating laser beams ($|\text{lin}, \perp \text{lin}|$). We attributed spatial modulation of the atomic potential energy to spatial modulation of the net field polarization. In a more general case, however, not only polarization of the net field is space-dependent, but also its intensity. This happens, for example, in a standard MOT configuration with three pairs of counter-propagating, $\sigma^+ - \sigma^-$ polarized beams. We can write a general expression for the potential energy of a sublevel $|F_g, m_g\rangle$ in the form

$$
U_{|F_g, m_g\rangle}(r) = U_{0,|F_g, m_g\rangle} s(r),
$$

(2.39)
where

$$s(r) = \frac{\Omega^2(r)}{\Delta^2 + \Gamma^2/4}$$

(2.40)

is the space-dependent saturation parameter and \(U_{0,|F_g,m_g\rangle}\) is the constant characterizing the light shift of the corresponding \(|F_g,m_g\rangle\) sublevel. Atoms travelling across periodic structure of the light potential experience both the Sisyphus sub-Doppler cooling and friction force due to the corkscrew standing wave [78, 52] and can eventually localize in potential minima of the optical lattice.

In principle, an optical lattice can be created by any superposition of several light beams. In particular, in a standard MOT, six intersecting trapping beams can interfere to produce periodic optical potential. Still, creation of an optical lattice in a standard MOT is not obvious. The difficulties in creating it are caused by the following circumstances. The net laser field necessary for tight confinement of atoms in a periodic structure should provide efficient Sisyphus cooling and circular polarization of light in the potential minima. This field, which is nothing else but the modulation of light intensity and polarization due to interference of the incident beams, depends strongly on their relative phases. It has been shown in Ref. [79] that when \(N\)-dimensional lattice is produced by \(N + 1\) laser beams, the relative phase shift results only in translation of the whole lattice structure without changing its topological properties. If more than \(N + 1\) beams are used to create \(N\)-dimensional lattice, their relative phase shift alters the lattice topology and can lead to less efficient atomic confinement. Thus, obtaining the desired topology of an optical lattice requires proper phase stabilization. In the case of a MOT, six beams are responsible for creating the polarization and intensity spatial modulation, thereby any phase fluctuations change the net field structure. Even if within finite time intervals the relative phase remains stable, its fluctuations and the resulting variations of the lattice topology hinder observation of stable vibrational spectra.

Moreover, a relatively small detuning from the resonance in a working MOT makes it difficult to confine atoms in optical potentials: since the photon scattering rate is proportional to \(I/\Delta^2\) and optical potential depth to \(I/\Delta\) \((I\) being the intensity of the trapping field), small detuning is not favorable for obtaining optical lattices.

Despite these difficulties, Schadwinkel et al. [81] managed to create optical lattice in a standard MOT by controlling the relative phases of the beams in appropriate setup [82]. It is very unlikely that in a MOT without special phase control a stable optical lattice field can be created.

### 2.9 Conclusions

Recoil-induced resonances, Raman resonances connected with vibrational levels in optical lattices and light-shifted Zeeman sublevels, discussed in Chapter 1, occur when the probe beam frequency is close to the frequency of the pump field. If no special techniques are used to isolate a given contribution, they all appear as a complex, subnaturally narrow resonant structure both in the probe absorption and four-wave mixing spectra. Different amplitudes, widths and different sensitivity of the resonances to the experimental conditions (like polarization of the probe beam, intensity of the pump field, etc.) allow one to treat the spectra as a diagnostic tool.

\[\text{This was the case in Ref. [80] where two-dimensional lattice was created using four beams}\]

\[\text{The pump field in the experiment described in this thesis is produced by the trapping beams.}\]
for a cold atomic sample. Positions of the Raman resonances between light shifted sublevels could be used for determination of the average Rabi frequency of the net pump field, whereas recoil-induced resonances give information about velocity distribution of the cold atomic sample (provided atoms are not bound in the direction of momentum exchange). The level of complexity of the resultant spectra, however, often makes the appropriate analysis difficult and sometimes ambiguous.
Chapter 3

Experimental setup

3.1 Introduction

The principles of laser cooling and trapping are extensively covered by vast literature (e.g. [17]-[19] and references therein). Thus, rather than describing physics connected with these processes we shall concentrate on description of the experimental setup used in this thesis.

Figure 3.1: Classical idea of a MOT. Three pairs of counter-propagating, orthogonally circularly polarized laser beams intersect in the region of trapping. A 3D magnetic field gradient is generated by a pair of coils with opposite currents.

The classical scheme of a magneto-optical trap, such as used in our experiment, is presented in [83]. The standard layout of a MOT is depicted in Figure 3.1. In brief, in order to cool and trap atoms three pairs of intense, counter-propagating laser beams
of orthogonal, circular polarizations ($\sigma^+ - \sigma^-$ configuration) are required together with magnetic field $B$ that zeroes at the trap center and has non-zero gradient along each direction. Such a configuration of $B$ is produced by magnetic coils in an anti-Helmholtz arrangement, i.e., with opposite electric currents in each coil. Since in the case of alkali metals the cooling process results in optical pumping of atoms to the hyper-fine state that is non resonant with the trapping laser, additional laser is required to bring them back to the state which is used in the trapping transition. To probe the atoms in a MOT we use another laser that is locked to the trapping one. Despite the relatively simple principle of cooling and trapping, experimental realization of the trapping fields requires fairly complex optical and electronic setup. Nevertheless, the MOT has already become a standard device for spectroscopy, widely used and technically mastered.

The magneto-optical trap described in this chapter evolved from previous setups of the lab, described in details in Refs. [84]-[86]. The present experimental setup is an optimized version of the one depicted in Ref. [86].

3.2 Lasers

In the experiment we use four diode lasers working at 780 nm:

- **MASTER LASER** (master oscillator), equipped with the SANYO 80 mW laser diode, model DL7140-201 [87] and external resonator formed by a holographic diffraction grating mounted in the Littrow configuration [88]. This laser is actively stabilized to the saturated absorption frequency reference [84, 86]. The dispersion-shaped error signal is derived from the absorption spectroscopy using a lock-in amplifier [89]. The frequency of the master laser is modulated by applying a sinusoidal signal (2 mVpp, 6 kHz) to the piezo transducer which changes the resonator length. The modulation amplitude corresponds to the laser frequency deviation of the order of $0.05\Gamma$ and was optimized to give a good error signal without disturbing the spectral properties of the laser too much. The master laser beam is used to synchronize the trapping and probe lasers. Details of the master laser design are described in Ref. [90].

- **REPUMPING LASER**, based on the HITACHI 50 mW laser diode, model HL7851G [91], working in a free-running mode. This laser is only current- and temperature-stabilized. The quality of this stabilization is sufficient to use the laser without external cavity for the repumping transition\(^1\). The repumping beam is mixed with one of the trapping beams.

- **PROBE LASER**, based on the HITACHII 50 mW laser diode, model HL7851G [91], is injection-seeded by the master laser beam and reproduces the spectral characteristics of the injected light.

- **TRAPPING LASER**, using the SANYO 80 mW laser diode, model DL7140-201 [87], is also injection-seeded by the master laser beam, thereby it is locked to the master and probe lasers. It offers substantial amplification of the master beam and powers up to 80 mW. In our experiment, the trapping beams play the role of pump beams. Due to the locking of the master, pump and trapping

\(^1\)This was verified by a long-term measurement of atomic fluorescence from the trap.
3.3 Laser frequency synchronization

The scheme of phase synchronization of lasers in our experiment with use of the injection-locking technique [92, 93] is presented in Figure 3.2.

Master laser

The beam from the master laser is divided by a beamsplitter cube. One part of the beam is used for stabilization of the master laser frequency as described in the previous section. Before entering the rubidium cell for saturated spectroscopy, the beam is frequency shifted in a double pass through an acousto-optic modulator.
AOM-0\textsuperscript{2}. We utilize the −1 order of diffraction on AOM-0, so the light that enters the cell has frequency

\[ f_{\text{sat}} = f_{\text{master}} - 2f_{\text{AOM-0}}, \]  

(3.1)

where \( f_{\text{master}} \) is the master laser frequency, \( f_{\text{AOM-1}} \) is the modulation frequency of AOM-1. We stabilize the master laser to the crossover resonance \( co(2-4) \) between the \( F = 3 \leftrightarrow F' = 2 \) and \( F = 3 \leftrightarrow F' = 4 \) transitions (see Figures 3.3 and 3.4), hence the master laser frequency becomes

\[ f_{\text{master}} = f_{\text{co}(2-4)} + 2f_{\text{AOM-0}}, \]  

(3.2)

where \( f_{\text{co}(2-4)} \) is the \( co(2-4) \) frequency. AOM-0 is used to change the detuning of the trapping beams from the atomic resonances (as shown later in this section). The double pass setup of the AOM results not only in a double frequency shift but also eliminates the beam deflection when the AOM driving frequency is varied.

The second part of the split master laser beam is used to seed the trapping and probe lasers.

**Trapping laser**

The trapping laser is injected with the master laser beam of frequency \( f_{\text{master}} \). The trapping beam is then frequency shifted by a single passage through AOM-3 adjusted for the −1 diffraction order\textsuperscript{3}. Thus, the trapping beam frequency equals

\[ f_{\text{trap}} = f_{\text{master}} - f_{\text{AOM-3}}. \]  

(3.3)

The crossover frequency \( f_{\text{co}(2-4)} \) differs from frequency \( f_{34} \) of the \( F = 3 \leftrightarrow F' = 4 \) transition by 92 MHz,

\[ f_{\text{co}(2-4)} = f_{34} - 92 \text{ MHz}, \]  

(3.4)

hence, using (3.2), (3.3) and (3.4) we can calculate the trapping beam detuning \( \Delta/2\pi = f_{\text{trap}} - f_{34} \) from the atomic resonance,

\[ \frac{\Delta}{2\pi} = 2f_{\text{AOM-0}} - f_{\text{AOM-3}} - 92 \text{ MHz}. \]  

(3.5)

The frequency \( f_{\text{AOM-3}} \) is set to 80 MHz. We change \( \Delta \) (typically between \( -\Gamma \) and \( -5\Gamma \)) by varying frequency \( f_{\text{AOM-0}} \) using a home-made, VCO-based controller [96].

**Probe laser**

Before injection of the master beam to the probe laser, it double-passes AOM-1 (here again we use -1 diffraction order). Thus, the probe laser is forced to oscillate at the frequency

\[ f_{\text{probe}} = f_{\text{master}} - 2f_{\text{AOM-1}}. \]  

(3.6)

AOM-1 is driven by the VCO-based controller of the same type as used for AOM-0. We use AOM-1 to sweep the probe beam frequency during spectroscopic measurements. The frequency sweep is realized by a triangular voltage ramp sent to the

\textsuperscript{2}All acousto-optic modulators used in the experiment are ISOMET 1205C with center frequency \( f = 80 \) MHz [94]. Except for AOM-2, they all are powered by the Mini-Circuits ZHL-3A RF amplifiers [95].

\textsuperscript{3}AOM-3 is driven by the HP 8647A frequency synthesizer.
3.3. LASER FREQUENCY SYNCHRONIZATION

Figure 3.3: Hyperfine structure of the $^{85}\text{Rb} D_2$ line ($\lambda = 780.027$ nm) with positions of cross-over resonances.

Figure 3.4: Saturation spectroscopy of the $^{85}\text{Rb} D_2$ line. Laser frequencies are indicated in boxes, arrows represent frequency shifts introduced by AOMs (see text).
modulation input of the VCO controller designed and manufactured by Michał Zawada [96]. The ramp is derived from the Agilent 33120A function generator. Finally, before entering the atomic cloud, the probe beam is frequency shifted by AOM-2⁴ (this time +1 diffraction order is used) close to \( f_{\text{trap}} \). The difference between frequencies of the probe and trapping beams, or, in the other words the probe-pump detuning \( \frac{\delta}{2\pi} \), equals

\[
\frac{\delta}{2\pi} = f_{\text{probe}} - f_{\text{trap}} = f_{\text{AOM-2}} + f_{\text{AOM-3}} - 2f_{\text{AOM-1}}.
\]

(3.7)

The precision reached in setting the \( \delta \) value, crucial for the measurements presented here, is assessed in Appendix B. The diagram of all AOM connections is depicted in Figure 3.5.

**Repumping laser**

The frequency of repumping laser is tuned to the \( F = 2 \leftrightarrow F = 3 \) trapping transition (see Figures 3.3 and 3.4). Since the requirements for spectral width of the repumping light are not as stringent as for other lasers⁵, the laser works without an external resonator and we do not actively stabilize its frequency. The precision of current and temperature stabilization is high enough to keep the laser on the desired line for long time. We monitor the proper tuning of the repumping laser frequency by observing fluorescence from the atomic cloud. As this fluorescence depends on the number of trapped atoms, it gives information about efficiency of the repumping process.

![Diagram of the AOM connections. Functions of the relevant AOMs are indicated in Figure 3.2.](image)

Figure 3.5: Diagram of the AOM connections. Functions of the relevant AOMs are indicated in Figure 3.2.

### 3.4 Magnetic field

The quadrupole magnetic field \( \mathbf{B} \) is generated by a pair of coils in the anti-Helmholtz arrangement (Figure 3.6). The coils used in the experiment have diameter \( 2R = 8 \)

---

⁴AOM-2 is controlled by the ISOMET 232A RF Analog Driver [97].

⁵The only role of the repumping laser is to depopulate the \( F = 2 \) hyperfine level.
cm and are separated by a distance \(2A = 5\) cm. Such a geometrical configuration \((2A = \frac{5}{4}R)\) assures constant radial \(\frac{\partial B}{\partial \rho}\) and axial \(\frac{\partial B}{\partial z}\) gradients of the magnetic field at the trap center \((±2\) mm\) and the same value of the field barrier in the axial and radial directions. Another possible coil arrangement, with \(2A = \sqrt{3}R\), offers a wider range of constant gradient but a lower value that can be achieved in the \(2A = \frac{5}{4}R\) case.

Figure 3.6: Magnetic coil configuration used in the experiment. Central round coils generate the quadrupole magnetic field for MOT. Three pairs of rectangular Helmholtz type coils are used to compensate stray DC magnetic fields. Directions of currents in the coils are indicated by arrows.

Each coil has 80 turns of 1 mm copper wire and carries current \(I_{\text{coil}}\) between 0.5 A and 5.5 A, depending on the desired gradient. For \(I_{\text{coil}} = 3\) A, the axial field gradient is \(\frac{\partial B}{\partial z} = 16\) Gauss/cm (radial field gradient is two times smaller\(^6\)). With a coil resistance of 1.8 Ω, we have a 4.5 V voltage drop at \(I_{\text{coil}} = 2.5\) A which yields 11 W of dissipated electric power. Although our coils are equipped with a water cooling system, we have not used it since the heat was sufficiently dissipated by air.

Stray DC magnetic fields, both the Earth’s and ion pump fields, add to the quadrupole magnetic field and spatially shift its zero point in an uncontrolled way. Since the trapping beams have Gaussian intensity profiles, the atom trapping can thus occur in a region of unbalanced radiation pressures. This fact could be observed after switching-off the quadrupole magnetic field. A fast opening of the trap results in a quite asymmetric, jet-like escape of atoms from the trapping region rather than in an isotropic expansion of the cloud in optical molasses. Such a behavior is

\(^6\)This is the consequence of \(\nabla \cdot \mathbf{B} = 0\) with \(\partial_x B_x = \partial_y B_y\).
undesirable, especially for the TOF measurements [98], as it disturbs the atomic free fall in the gravitational field.

In order to circumvent the problem of stray DC magnetic fields, a set of three pairs of Helmholtz-type compensation coils (Figure 3.6) is used. These coils generate a constant bias magnetic field in the trapping region. The compensation of stray DC fields is based on observation of the evolution of the atomic cloud after the quadrupole field is turned-off. By changing the current running through the relevant coils, we are able to achieve the isotropic cloud expansion, which is the evidence for a perfect stray magnetic field compensation (Figure 3.7).

More detailed description of the coils used in our experiment can be found in Ref. [86].

Figure 3.7: Expansion of a cold atomic cloud in optical molasses. Quadrupole magnetic field was switched-off at time \( t = 0 \) (first frame), repumping and trapping beams remained turned-on. The cloud expands isotropically, which is a signature of well compensated stray DC magnetic fields in the center of the trap. Without such compensation the cloud is abruptly pushed away of the trap due to imbalanced radiation pressure. In the first frame \( g \) shows direction of the gravity.

3.5 Vacuum chamber

We trap atoms in a vacuum chamber initially pumped by a turbo-molecular pump (Balzers/Pfeiffer TPH170) then by a titanium differential ion pump (Physical Electronics 12S) with pumping speed 12 l/s, controlled by the Physical Electronics IONPAK driver. The ion pump remains switched-on during the whole experiment, which yields a vacuum chamber pressure below \( 10^{-9} \) mbar.

The vacuum apparatus setup is depicted in Figure 3.8. Optical access to the stainless steel chamber is provided by glass windows (3.5 cm in diameter) with
3.5. VACUUM CHAMBER

Figure 3.8: Vacuum chamber setup.
antireflection coating (780 nm, 0°). Beneath the main part of the chamber, 8 cm below the MOT center, we placed small windows for introducing a probe beam for the time-of-flight temperature measurements. The quadrupole magnetic coils are attached to the vacuum apparatus which assures their central position with respect to the z axis.

The rubidium source for our trap is the alkali metal dispenser (AMD, [99]), often called a getter. Release of rubidium atoms occurs when the dispenser is heated in vacuum, which starts chemical reactions leading to metal evaporation. Dispenser heating is realized electrically. Usually, current of the order of 3 A is needed to keep a constant Rb vapor pressure. After a longer break in using the getter source, one needs to refresh it by applying short pulses of 5-6A. The advantage of using a dispenser over an oven, the standard atom source in most MOTs, is the ease of use, better control and far smaller inertia in the number of atoms in the vacuum chamber. Properties of a getter source for a MOT are described in detail in Ref. [100].

3.6 Optical setup

The layout of optical setup is presented in Figure 3.9. It shows the actual beam guiding up to the vacuum chamber, which is shown here only schematically. Most of important features of the setup have been already described in Sections 3.2 and 3.3. The remaining parts of the setup are briefly discussed below.

Saturation spectroscopy detectors

Signals from the saturation spectroscopy are monitored with the \( \text{phd (m)} \) and \( \text{phd (r)} \) detectors for the master and repumping lasers, respectively. These detectors are based on the Hamamatsu S2386-44k photodiode [101]. Since they are utilized in a phase-sensitive detection, their bandwidth should be broad enough to detect the 6 kHz modulation of the incident signal. The detectors were designed and manufactured by Tadeusz Pałasz. Their circuit diagrams and characteristics can be found in Ref. [84].

Optical isolators

Optical isolators\(^7\) (\( \text{io} \)) are used to reduce the influence of parasitic reflections on the laser generation. Moreover, we use additional ports of two isolators to inject the master laser beam into the trapping and probe lasers as shown in Figure 3.9.

Beam matching in the injection locking

Injection locking works efficiently when the injected beam and laser output are matched geometrically: they should have the same size and divergence, which is difficult to achieve. We were able to lock the injected lasers very efficiently by focusing the master laser beam with lenses placed before the additional ports of \( \text{io} \). The focal lengths of the lenses were adjusted empirically for the best injection efficiency, which was much simpler than shaping the beam to match the laser output. This method resulted in very good injection quality even when the unlocked slave laser was far detuned from the master laser.

\(^7\)Isowave I-80-T5H, 40dB.
Figure 3.9: Optical setup of the experiment.
Laser diagnostics

Spectral properties of all lasers are monitored by a spectrograph\textsuperscript{8} (SG) and confocal Fabry-Perot spectrum analyzer\textsuperscript{9} (F-P). Lasers are coupled to the spectrograph by optical fibers. The light is taken either from spare reflections, e.g. from prisms, or unused 0-th AOM diffraction order. The spectrograph is used for coarse analysis of the laser wavelength and is particularly useful when lasers are far detuned from the desired transitions. The scanned Fabry-Perot spectrum analyzer gives information about the mode structure of laser radiation and allows one to test quality of the injection locking. This is done by observing how the frequency of the injected laser follows the modulation of the master laser frequency. Our laser diagnostics setup is completed by saturation spectroscopy of Rb in a glass cell that has to be performed before the master laser is locked to the atomic resonance. Very helpful for the laser tuning is the infrared camera that monitors fluorescence from two rubidium vapor cells used in saturation spectroscopy.

Shaping of laser beams

Laser beams are first geometrically shaped by anamorphic prisms which correct their elliptical profile to the circular one. The prisms can be set either for expansion or contraction of the beam profile. Additionally we use two $f = 100$ mm lenses in a 1:1 telescope setup\textsuperscript{10} for suppression of the trapping beam astigmatism. This is done by a slight rotation of the lens around the axis perpendicular to the beam axis. Repumping and trapping laser beams are then expanded by magnifying telescopes and reach about 1 cm in diameter. The trapping beam is spatially filtered by a $50 \mu m$ pinhole placed in the focus of the expanding telescope. After the expansion of the trapping beam we have about $500 \mu W/mm^2$ of its maximum intensity at our disposal.

Measurement of laser beam intensity

For the measurements of laser beam intensities we use the Coherent LaserMate-Q powermeter with calibrated 1 mm\textsuperscript{2} iris mounted on its VIS sensor. All intensity measurements in this thesis refer to the maxima of the beam intensity distributions. Additionally, we use the Large Active Area ($10 \times 10$ mm) Hamamatsu S2387 photodiodes [101], calibrated with the powermeter and connected directly to a digital ammeter\textsuperscript{11}. Usually, it is sufficient to illuminate photodiodes with light that leaks through the mirrors or with spare reflections. Thus they can be placed out of the beam paths, which makes them a very comfortable tool for the on-line beam power measurements.

\textsuperscript{8}Two-meter, plane grating spectrograph Carl Zeiss Jena PGS2, dispersion 3.55 Å mm\textsuperscript{-1}.
\textsuperscript{9}Cobrabid KB6316, FSR=1.5 GHz, $\delta = 100$.
\textsuperscript{10}One of the lenses is placed before the input of the trapping laser and the other at its exit.
\textsuperscript{11}Conrad DPM 95 Panel Meter.
3.7 Setup for pump-probe spectroscopy of trapped atoms

Experimental realization of the pump-probe spectroscopy with trapped atoms is depicted in Figure 3.10. Before entering the cloud, the probe beam is sent through a polarizing beamsplitter cube which assures its linear polarization and, together with the $\lambda/2$ waveplate, allows one to control the beam power. A $\lambda/4$ waveplate after the beamsplitter cube changes the probe beam polarization from linear to either $\sigma^+$ or $\sigma^-$. The closest trapping beam (indicated in Figure 3.10a by a grey stripe) is $\sigma^-$-polarized. The probe beam is now directed onto the atomic cloud by the edge mirror so that it does not block the trapping beam. The probe beam makes a small angle $\theta \approx 3^\circ - 7^\circ$ with the closest trapping beam.

![Figure 3.10: (a) Pump-probe spectroscopy setup. Filled and hollow arrows indicate propagation of the probe beam and FWM signal, respectively. (b) Data acquisition diagram.](image)

To introduce the probe beam into the cloud, we set its power to maximum, contract its spatial profile by an iris and align the guiding mirrors to obtain a dark spot in the center of the cloud (Figure 3.11). This dark spot is due to radiation pressure of the probe beam which pushes atoms out of the trap. After the probe beam passes the cloud, it is directed on the detector ABS phd. The four-wave mixing signal (FWM), simultaneously generated in the atomic cloud, is propagating in the direction opposite to the probe (hollow arrows in Figure 3.10a). It is reflected by the beamsplitter plate and directed onto highly sensitive detector FWM phd, described in the next Section. In this manner, the ABS and FWM signals are recorded simultaneously and directly, without any phase shifts that could possibly be introduced if one would use lock-in detection. To align the FWM phd, we first retroreflect the probe beam replacing the ABS phd with a mirror and then adjust position of FWM phd to find optimal signal from the reflected probe beam.$^{12}$

$^{12}$Such configuration allows also the ABS measurement with twice the optical path due to a
The scheme of data acquisition is very simple (Figure 3.10b). Both **ABS phd** and **FWM phd** are connected to the oscilloscope\(^{13}\), which is triggered by the function generator\(^{14}\) responsible for the probe frequency sweep. The recorded voltage ramp from this generator, sent to the input of AOM-1 VCO, is then used for calibration of the frequency axis. The acquired signals were averaged using the oscilloscope’s built-in mathematical functions. Usually, up to 30 traces of ABS and FWM signals were used for averaging.

Figure 3.11: Dark spot in atomic cloud. It is caused by a high intensity probe beam, which pushes atoms out of the cloud (due to uncompensated radiation pressure). This phenomenon was used to direct the probe beam precisely through the cloud center.

### 3.8 ABS and FWM detectors

The power of FWM signals generated in nonlinear processes in a MOT is of the order of several tens of nanowatts. To be able to measure such a weak signal directly without phase sensitive detection, a low-noise, high gain detector is needed. Since our sweep rates are not too fast (3 MHz/sec)\(^{15}\), the detector gain can be enhanced at the expense of its bandwidth.

Our FWM signal detector uses two amplification stages. The first one is based on operational amplifier integrated with photodiode, OPT101 [102]. With a 1MΩ resistance in a feedback loop of the amplifier we get 0.4 V/µW at 780 nm at the exit of OPT101. This signal has to be amplified at least 100 times. This is done by op-amp OP27 [103]. In our detector we achieve 30 mV/nW final amplification (it is slightly reduced due to an IR filter placed in front of OPT101). The detector has also to provide compensation of an offset signal, which is realized with the subcircuit based on a 2.5 V reference diode (LM336). The voltage drop across 10 kΩ potentiometer parallel to the diode is fed to the non-inverting input of OP27. This allows one to precisely null the detector offset when no light impinges on OPT101. The capacitors shown in the diagram are used for noise reduction. Characteristics of the FWM detector are presented in Figure 3.13 and show that the detector cannot be used for signals that change faster than 2 kHz.

The ABS detector is based on a standard application of the OPT101 described in Ref. [102]. Measured amplification of our ABS detector is 0.39 V/µW.

Both detectors are supplied from batteries to avoid pickup of the mains noise.

\(^{13}\)LeCroy 9361 Dual 300 MHz 2.5 GS/s Digital Oscilloscope.

\(^{14}\)Agilent 33120A Function Generator.

\(^{15}\)This is primarily due to a narrow sweep range, typically −3...3 MHz.
3.8. ABS AND FWM DETECTORS

Figure 3.12: Circuit diagram of the FWM detector. Capacitor $C$ can be used to reduce noise at the first amplification stage.

Figure 3.13: Measured characteristics of the FWM detector.
Chapter 4

Experimental results

4.1 Introduction

In Chapters 1 and 2 we discussed the possible nonlinear processes that influence absorption and four-wave mixing spectra of atoms subject to intense light fields. Chapter 1 is devoted to the effects connected with altering the energy levels of an atom by its interaction with light, while Chapter 2 refers to the spectroscopic manifestation of atomic motion in sufficiently cold samples.

In the experiment discussed in this thesis, we probed atoms in a magneto-optical trap. In our MOT, the temperature of the atomic sample is as low as 100 $\mu$K [98]. Such a narrowing of the atomic momentum distribution results, among other things, in a practical cancellation of the Doppler broadening. This greatly enhances the resolution of spectroscopic measurements. Moreover, in the regime of such low temperatures, atomic motion can be examined by means of spectroscopic methods. Thus, the movement of atoms is no longer a disturbing, undesired factor, but should be considered rather as an interesting phenomenon, influencing the spectra in a non-trivial way. Indeed, as it will be shown in the following sections, the spectra described in this thesis exhibit resonant contributions that have to be explained with inclusion of atomic motion.

The interpretation of the spectra presented below concentrates mainly on the absorption features. We also present simultaneously recorded four-wave mixing (FWM) signals. Many features of the FWM signals indicate significant contribution of the atomic motion. Since the FWM signals are characterized by better resolved resonances when compared with absorption, we strongly believe that they can constitute a powerful diagnostic tool for the processes that occur in cold atomic samples. Obviously, in order to take advantage of the FWM diagnostics, accurate theoretical treatment of the FWM in cold atoms should be available. The present theory [60]-[64] which incorporates the simple atomic level schemes and basic geometries of pump and probe beams, albeit complicated, describe much too idealized situations to offer full understanding of our experimental results connected with FWM.
4.2 Experimental configuration and basic terminology

In this section we concentrate on the principle of the experiment. Technical details connected with laser tuning and other features of the setup can be found in Chapter 3.

The experimental configuration is depicted in Figure 4.1. The probe beam makes a small angle $\theta$ (usually between 3° and 7°) with the $z$ axis, which is the symmetry axis of the quadrupole magnetic coils (the magnetic field gradient along $z$ is twice as big as in the radial direction). The trapping beam closest to the probe beam (nearly co-propagating with the probe beam) is $\sigma^-$-polarized. Hence, when the probe beam is $\sigma^+$-polarized, it has the same polarization as the oppositely propagating trapping beam. The angle between the two beams of the same polarization is then of course $(180° - \theta)$.

![Figure 4.1: Experimental configuration of the pump-probe spectroscopy of atoms in a MOT. Trapping beams play the role of pump beams, the probe beam propagates under a small angle $\theta$ to the one of them.](image)

When performing measurements with the trapping beams’ power as the variable parameter, the trapping beams usually have equal intensity (its peak value is about 120 $\mu$W/mm$^2$ at the maximum of beam Gaussian profile). In some measurements, however, we approach the 1D configuration by increasing the power of the beams propagating along the $z$ axis (the pair closest to the probe beam) at the expense of the remaining trapping beams. The pairs of trapping beams along $x$ and $y$ axes have thereby lower, but equal intensities. The intensity imbalance between radial ($x, y$) and axial ($z$) directions result in cloud compression along $z$. This procedure allows approximation of the 1D situation while maintaining a stable trap and allows its study in stationary conditions.

We register the probe transmission signals, which we call “absorption spectra” (ABS) and simultaneously the four-wave mixing signals (FWM) as a function of
4.3 WIDE-SCAN SPECTRA

the pump-probe detuning $\delta$. The scan of $\delta$ is performed in two regimes: wide and narrow. *Wide scan* refers to the case when $\delta$ is swept within limits of $-40 \ldots +40$ MHz ($-6.7\Gamma \ldots 6.7\Gamma$). The range of the sweep allows one to examine the resonances associated with dressing of atom by photons of the trapping beams (see Section 1.4, page 13). These resonances are called the *Rabi sidebands*. Depending on the sign we distinguish the *gain sideband* or *absorption sideband*. Sidebands are observed for $\delta$ of the order of a few $\Gamma$. As it can be seen from Figure 4.2, the spectra exhibit very narrow resonances for $\delta \approx 0$. These resonances are the main subject of this thesis and are studied experimentally in the *narrow-scan* regime, for which $\delta$ is swept by $\pm 2.5$ MHz ($\pm 0.4\Gamma$) around zero. The resonances observed for such values of $\delta$ constitute the so-called *central structure* (CS). According to considerations from Chapters 1 and 2 the central structure is a net result of the Raman transitions between Light-Shifted Sublevels (RLSS) of the DTLS, recoil-induced resonances (RIR) and resonances due to the Raman transitions between Vibrational levels in Optical Lattices (RVOL).

Figure 4.2: Examples of experimental absorption and four-wave mixing spectra recorded in wide and narrow scans of the pump-probe beam detuning $\delta$. For these spectra, the probe is $\sigma^+$-polarized, intensity of the trapping beams is $I = 105 \mu W/mm^2$ per beam (measured at the maximum of the beam gaussian profile), trapping beam detuning from resonance $\Delta = -3\Gamma$, and axial magnetic field gradient $\frac{\partial B}{\partial z} = 13$ Gauss/cm.

4.3 Wide-scan spectra

The wide-sweep regime allows one to probe the energy structure of an atom dressed by photons of the trapping beams. According to discussion in Sections 1.4 and 1.5, we expect the ABS and FWM signals to have sideband resonances for pump-probe
Figure 4.3: Set of the ABS spectra for increasing trapping beam intensity. $I$ stands for the maximum intensity of one of the trapping beams, $\Delta = -3.1 \Gamma$, probe polarization is $\sigma^-$, magnetic field gradient $\frac{\partial B}{\partial z} = 13$ Gauss/cm. The dotted line marks the position of the unperturbed atomic resonance ($F = 3 \leftrightarrow F' = 4$). All plots are in the same scale. Inset shows dependence of the sideband positions on the trapping beam intensity (per one beam). The solid line is a function $\delta_{\text{res}} = \pm \sqrt{\Omega^2 + \Delta^2}$, where $\Omega^2 = rI$. The scaling factor $r = 0.4$ was adjusted to reach the possible good agreement with both gain and absorption sideband positions. Despite quantitative agreement, the differences between the simple theory (Figure 1.7, page 16) and experiment are well visible.
detuning $\delta = \pm \Omega'$ and some resonant structure for $\delta = 0$. Since the trapping beam detuning $\Delta$ from the atomic resonance is negative (typically $\Delta = -3\Gamma$), the ABS signal should show gain for $\delta = -\Omega$ and attenuation for $\delta = \Omega$. Indeed, the wide scan plots in Figure 4.2 exhibit all expected features. Increase of the trapping beam power and the corresponding growth of $\Omega'$ result in the shift of the position of the resonances away from the center of the spectra. Figure 4.3 shows this effect in the case of ABS spectra.

By closer inspection of the wide-scan spectra depicted in Figures 4.2 and 4.3, however, one notices many qualitative and quantitative departures from the expectations based on the simplistic model of the two-level system:

- the form of central structure in both ABS and FWM spectra;
- the width and shape of resonances;
- the asymmetry in positions of the gain and absorption sidebands;
- different positions of the sideband resonances in the simultaneously registered ABS and FWM spectra.

Now, we shall explain these features of wide-scan spectra.

### 4.3.1 Central structure

It was mentioned already in Section 1.13 that only some special configurations of pump-probe field polarization allow one to treat the DTLS as a two-level system (or a set of two-level systems). Rubidium atoms in the trap are subject to the net trapping field of a complex, spatially varying polarization structure (Appendix D), hence in most cases the DTLS structure with RLSS processes has to be taken into account. This results in appearance of the specific central resonant structure around $\delta = 0$, as it was discussed in Section 1.14. Moreover, for typical MOT temperatures one has to consider the resonant contributions associated with photon recoil and atomic localization (the RIR and RVOL processes), which occur also for the small probe-pump detuning. The central structure will be analyzed in detail further in this Chapter.

### 4.3.2 Width, shape and position of the sidebands in ABS and FWM spectra

Inclusion of the DTLS structure in calculations of the ABS and FWM signals results not only in appearance of the CS but also broadens resonances with respect to the case of a two-level system. This is due to different Rabi frequencies between different sublevels and specific population distribution (alignment). This effect has been shown in case of $\pi - \pi$ pump-probe polarization configuration. However, this broadening is too small to explain the experimental results. Figure 4.4 shows this incompatibility of the experimental ABS spectrum with the numerical simulation [44] for corresponding experimental parameters and $\sigma-$-$\sigma$-$\pi$ pump-probe polarization configuration. Comparison of the plots in this Figure emphasizes differences in the shape and width between theoretical and experimental spectra. Moreover, positions of the sideband resonances in numerically generated DTLS FWM spectra coincide with those in the ABS spectra, which is not the case for an experimental results.
discrepancies motivate us to look for explanation of the observed disagreement and extension of the DTLS model to describe realistic conditions of trapped atoms.

Numerical simulations of the DTLS spectra such as shown in Figure 4.4 are performed for the well-defined pump-probe polarization configuration and constant value of the Rabi frequency $\Omega$. Neither of this two conditions, however, is fulfilled in the trap. The interference of six Gaussian beams results in the spatial pattern of periodic light and polarization modulation (see Appendix D and Ref. [85]). Thus, the Rabi frequency and pump-probe polarization configuration are position-dependent. Atoms, depending on their position, experience various schemes and strengths of excitation by the net trapping field. Periodicity of the modulation guarantees that a moving atom experiences average values of the corresponding net field parameters. Since the net trapping field modulation depends on the relative phases of the trapping beams, the processes of light-atom interactions become averaged not only by the atomic movement but also by mechanical fluctuations of the experimental setup.

Although the situation appears very complicated, the spectra obtained in the experiment are very stable and reproducible with distinct systematic dependencies of the resonance positions, widths and amplitudes on experimental conditions. This encourages one to try a simple modelling of the ABS and FWM signals by generating the DTLS spectra for certain ranges of Rabi frequencies and different pump-probe polarization configurations. If the hypothesis that light and polarization modulation is indeed the main reason of the observed shapes of the ABS and FWM signals, the weighted sum of the numerically generated lineshapes should produce good agreement with the experimental spectra.

Another relevant condition is a finite optical thickness of the medium. Absorption sidebands registered in the experiment are associated with a 60-70% attenuation of the probe beam power. It means that rather than plotting absorption coefficient
4.3. WIDE-SCAN SPECTRA

\( \alpha(\delta) \), which is a standard approach with optically thin media, we shall use the transmitted intensity calculated from the Beer’s law, namely

\[
I(\delta) = I_0 e^{-\alpha(\delta)},
\]

where \( I(\delta) \) is the intensity of the probe past the medium, \( I_0 \) is the incident intensity of the probe and \( c \) is a constant responsible for optical thickness of the medium. Of course, when \( \alpha(\delta) \ll 1 \) (the case of optically thin medium), we have

\[
I(\delta) \approx I_0 [1 - \alpha(\delta)].
\]

To verify whether such effects of field inhomogeneity can explain the experimental results, we have generated the DTLS spectra for the range of Rabi frequencies \( \Omega \in [0; 8\Gamma] \) and for various pump-probe polarization configurations: \( \pi - \sigma \pi, \sigma - \sigma \pi, \sigma^+ - \sigma^+ \) and \( \sigma^- - \sigma^+ \). We expect that the choice of such configurations covers a wide range of polarization schemes that occur in a MOT. For example, interference of two orthogonally circularly polarized counter-propagating beams results in a linearly polarized net field with spatially modulated direction. If we take direction of the \( E \) field as a local quantization axis, the pump field becomes \( \pi \)-polarized and a circularly polarized probe can induce \( \sigma \) and \( \pi \) transitions. The \( \sigma^+ - \sigma^+ \) pump-probe polarization configuration is responsible for a pure two-level contribution to the net signal. The \( \sigma^- - \sigma^+ \) configuration should also be taken into account, due to the symmetry of the setup. The spectra corresponding to chosen basic polarization configurations have certain features\(^1\) consistent with the actual experimental signal, which helped us in making the discussed selection.

Since the polarization and intensity modulation occurs with 100% contrast only in specific combinations of the trapping beams phases and each polarization configuration can be characterized by its own modulation pattern, we added the polarizations configurations averaged over Rabi frequency ranges and weighted them as follows:

- \( \pi - \sigma \pi: \Omega \in [5.1\Gamma; 7.3\Gamma] \), weight 1;
- \( \sigma - \sigma \pi: \Omega \in [3.1\Gamma; 6.2\Gamma] \), weight 0.96;
- \( \sigma^+ - \sigma^+: \Omega \in [0.7\Gamma; 3.7\Gamma] \), weight 0.41;
- \( \sigma^- - \sigma^+: \Omega \in [0.7\Gamma; 3.7\Gamma] \), weight 0.41.

These ranges and weights correspond to the conditions realized in a real trap with trapping beams of arbitrary phases.

The resultant ABS spectrum, plotted in Figure 4.5 using the Beer’s law (4.1), shows a very good agreement with the experimental spectrum. For exactly the same Rabi frequency ranges and weights as in the case of the ABS spectrum, we have also plotted the FWM spectrum. As it can be seen, agreement of the sideband positions between experimental and theoretical spectra is also very good, apart from a small shift of the sideband for \( \delta > 0 \). This shift can possibly originate from the attenuation of the probe beam around the center of the absorption sideband, i.e. from the decrease of the FWM power generated in a given frequency range.

\(^1\)For example, the gain sideband amplitude, the ration of the CS and absorption sideband amplitudes, shapes of the sidebands, etc.
What is more important, however, is that the averaging procedure (the same as in the case of ABS spectrum) produced FWM signal with sidebands having frequencies different than in the case of absorption. This is due to the fact that the $\sigma^+ - \sigma^+$ pump-probe polarization contribution, which pulls the whole absorption sideband in the ABS spectrum towards $\delta = 0$, does participate in the FWM signal as the dominant component shifting the sidebands away from $\delta = 0$. This behavior is caused by averaging over a certain range of the Rabi frequencies.

Although the agreement between theoretical predictions and experimental data is good, some discrepancies remain. First, the theory fails in reproducing the CS of the spectra. Second, there is a certain departure of the ABS theoretical curve from the experimental data for $\delta \in (0; 2\Gamma)$.

In order to verify whether the power of the probe beam affects the recorded lineshapes, we have performed a series of measurements with increasing probe power (Figure 4.6). The slopes of the absorption sideband remained essentially unaltered.

---

2Similar ABS spectra were recorded in Refs. [104],[105] and [49], but no satisfactory detailed explanation of their shapes was presented.
Figure 4.6: Influence of the probe beam intensity on the ABS spectra. A visible distorsion of the absorption sidebands is caused by the probe-beam radiation pressure pushing atoms out of the trap for $\delta$ around the absorption sideband center ($\delta \approx 4.3\Gamma$). Beyond this region the spectra remain unchanged. Experimental conditions: trapping beam power $I=253\ \mu W/mm^2$ per beam, detuning $\Delta = -3\Gamma$, $\pi$-polarized probe beam, axial magnetic field gradient $\frac{\partial B}{\partial z} = 13$ Gauss/cm.
even for high probe intensities. The main effect of the finite probe power is the pushing of atoms out of the trap for the probe frequency close to the center of the absorption sideband. Thus, the discussed differences between theory and experiment cannot be attributed to the probe beam power.

4.3.3 Conclusions

The wide-scan ABS and FWM spectra exhibit structures which qualitatively agree with predictions of the dressed atom model. However, in order to reach a satisfactory agreement between the experiment and theory, one has to employ the DTLS description of the medium and take into account the light intensity and polarization modulation in a MOT. We have shown that by averaging the DTLS spectra over certain Rabi frequency range and by taking into consideration various polarization configurations one can reproduce shapes of the ABS and FWM signals. Although some differences between theory and experiment require further studies, we have found the main reasons of spectral broadening and explained shifts of the sideband positions in of the FWM vs. ABS spectra.

Analysis of the positions of Rabi sideband resonances in a wide-frequency range has been previously performed in Ref. [84].

4.4 Narrow scan spectra

4.4.1 Introduction

Narrow-scan spectra are registered for probe-pump detuning of the order of a fraction of the natural linewidth, which for the $^{85}$Rb $5^2S_{1/2} \leftrightarrow 5^2P_{3/2}$ transition is $\Gamma = 2\pi \cdot 5.98$ MHz. Actually, we observe resonant structures that are not only well below $\Gamma$ but also below the laser linewidth, which in our experiment is about 1 MHz. As it is shown experimentally in Appendix B, accuracy of the relative pump-probe frequency determination is at least 3 kHz. This is due to the fact that the trapping and probe lasers are both optically coupled to one stabilized laser with external resonator (injection locking technique [92, 93], described in Chapter 3). Thus, all phase fluctuations causing laser line broadening are the same for these two lasers. Since all processes (RLSS, RIR and RVOL) responsible for resonances in the central structure (CS) are associated with Raman transitions, the phase fluctuations cancel out allowing one to resolve structures inaccessible by classical laser spectroscopy. A convincing comparison of the spectra obtained with and without coupling of the trapping and probe lasers can be found in Ref. [84]. Figure 4.7 shows the actual resolution that is reached in the present experiment.

In this section we make an attempt to interpret resonances in the narrow scan spectra obtained by probing the atoms in a working MOT. All the processes mentioned earlier can occur in the MOT conditions, hence we expect CS to be a complex superposition of RLSS, RIR and RVOL contributions. The situation is additionally complicated by the presence of the quadrupole magnetic field and by geometry of the net pump field, which is created by the interference of six trapping beams (Appendix D). To the best of our knowledge, all but one (Ref. [81]) previously reported study of the RLSS, RIR and RVOL has been performed with dedicated pump-probe configuration ([50, 51],[57]-[59], not to mention the vast literature on such studies with optical lattices.). In these experiments, the MOT was only used for preparation
4.4. NARROW SCAN SPECTRA

Figure 4.7: Example of the resolution capabilities of the present experiment. Numbers in the block arrows show magnification of the frequency scales in successive spectra due to decrease of the pump-probe detuning sweep range. We are able to resolve structures whose width is the kHz range.

of the cold sample, while spectroscopy was performed with extra pump and probe beams, of well-defined polarization and geometry, independent of MOT trapping field. Such a procedure greatly facilitated the interpretation of experimental results.

In this work, however, we use spectroscopic signals to acquire information on the trap conditions, so it is essential to record the signals while the trap is on. Explanation of the CS is greatly supported by the specific dependence of resonances associated with the discussed processes on the experimental parameters.

Raman transitions between light-shifted sublevels (RLSS)

The positions of resonances due to RLSS are governed by differential shifts of the ground state sublevels. Depending on the interaction strength, these shifts can be proportional to the light intensity \(I\) of the pump field (for \(\Omega \ll \Gamma, \Delta\)) or to \(\sqrt{I}\) (for \(\Omega \gg \Gamma, \Delta\)). Since the MOT operates mainly in the latter regime, we expect the \(\sqrt{I}\) resonance position dependence. Moreover, the quadrupole magnetic field, which zeroes only in the trap center, shifts Zeeman sublevels of an atom in dependence on its position in the cloud, causing inhomogeneous broadening of the RLSS resonances. Characteristic ratios of the positions of the \(|m_g = 0\rangle \leftrightarrow |m_g = \pm 1\rangle\), \(|m_g = \pm 1\rangle \leftrightarrow |m_g = \pm 2\rangle\) and \(|m_g = \pm 2\rangle - |m_g = \pm 3\rangle\) resonances, determined by the appropriate Clebsch-Gordan coefficients, can also help in identification of the resonances.

Recoil-induced resonances (RIR)

The positions of resonances due to atomic recoil depend on the temperature of the atomic sample and the geometry of the pump and probe beams. Thus, the RIRs depend indirectly on the trapping beam intensity \(I\) and detuning \(\Delta\). Decreasing \(I\) and increasing \(|\Delta|\) lowers the sample temperature, which in turn influences positions
of the RIR resonances. Also, by changing the angle between the pump and probe beams one can alter the width of the RIR structure. Both tests, however, can only partly be performed in our setup, where the angle \( \theta \) between the probe and the closest trapping beam is limited for technical reasons. Moreover, all MOT beams contribute to generation of the RIR signal. Thus, a change in the atomic temperature due to variations of \( I \) and \( \Delta \) can be too small to be measured with sufficient precision using the recorded spectra. Additionally, since the shape of the RIRs is a derivative of the velocity distribution (see Section 2.2), this distribution should be narrow enough (of the order of few \( \hbar k/m \)) in order to yield a measurable signal. Thus, the RIRs are observed only for the sufficiently cold samples.

Raman transitions between vibrational levels in optical lattices (RVOL)

Increase of the light intensity \( I \) should correspond to the increase of vibrational frequencies in optical lattice. On the other hand, when \( I \) grows, the temperature of the sample also increases which may prevent atomic localization. Moreover, the observed instability of the trapping beam phases (Appendix C) makes the hypothesis of optical lattices in our MOT very unlikely. That is why we should not regard the RVOL processes as a significant contribution to the ABS and FWM spectra, although complete absence of atomic localization in a MOT optical field remains yet to be proven.

4.4.2 Dependence of the CS resonances on the intensity of the trapping beams

The central structure has been investigated by changing intensities of the trapping beams and recording the ABS and FWM signals for the \( \sigma^+ \) and \( \sigma^- \)-polarized probe beam in the range of \( \pm 3 \) MHz around the trap beam frequency. The results are presented in Figures 4.8 and 4.9. In order to understand them we assume the model of DTLS perturbed by the \( \pi \)-polarized pump and probed by the circularly polarized probe, as defined in Figure 1.23, page 37.

In the ABS spectra we follow the behavior of resonances \( a, b \) and \( c \), as defined in the last plot in Figure 4.8. Brief inspection of the ABS spectra reveals the tendency characteristic for RLSS – the increase of trapping beam power results in a shift of the resonance positions from \( \delta = 0 \). Still, association of the observed resonances with RLSS meets several difficulties. First, if the \( a \) and \( b \) resonances are assigned to the transitions \( |m_g = 0\rangle \leftrightarrow |m_g = \pm 1\rangle \) (see Figure 1.23) and resonance \( c \) to the \( |m_g = 1\rangle \leftrightarrow |m_g = 2\rangle \) transition, we should expect the gain counterpart of resonance \( c \) on the red side of the spectra (for \( \delta < 0 \)). The only sign of this resonance is a broad and small bump that can be barely noticed in the \( \sigma^- \) spectra for \( I \approx 89.9 \mu \text{W/mm}^2 \) and \( I \approx 102.7 \mu \text{W/mm}^2 \). Second, there are no resonances associated with the \( |m_g = \pm 2\rangle \leftrightarrow |m_g = \pm 3\rangle \) transitions. We could expect that these resonances are so broad that they merge with the \( c \) resonance and its counterpart at \( \delta < 0 \). Should this be true, the \( |m_g = \pm 2\rangle \leftrightarrow |m_g = \pm 3\rangle \) resonances would have to be much wider than \( c \). This provokes the question: what is the reason for the different width of the RLSS resonances, already exemplified by the \( a \) and \( c \) resonances?

Despite doubts connected with the assumed model, we have plotted the positions of resonances \( a, b \) and \( c \) versus the trapping beams intensity (Figure 4.10). The
Figure 4.8: Series of the ABS spectra recorded for various trapping beam intensities for $\sigma^+$ and $\sigma^-$-polarized probe beams. Intensity $I$ per one beam is indicated beside every plot. Trapping beam detuning $\Delta = -3\Gamma$, axial magnetic field gradient $\frac{\partial B}{\partial z} = 13$ Gauss/cm. Central, ultra-narrow structure is marked in grey on the last plot, as well as the designation of resonances $a$, $b$ and $c$. 
Figure 4.9: Series of the FWM spectra – experimental conditions are the same as in the previous Figure.
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Experimental data is fitted with the function

\[ f_{ab}(\Omega) = \frac{1}{2} \sqrt{|c g_i^2 \Omega^2 + (\Delta + \frac{i}{2} \Gamma)^2|} \cos \frac{\phi_i}{2} - \frac{1}{2} \sqrt{|c g_i^2 \Omega^2 + (\Delta + \frac{i}{2} \Gamma)^2|} \cos \frac{\phi_i}{2}, \quad (4.3) \]

where \( c g_i \) is the Clebsh-Gordan coefficient associated with the \( \pi \) transition starting from the \( |m_g = i\rangle \) sublevel and \( \phi_i \) is the phase of \( z_i = c g_i^2 \Omega^2 + (\Delta + \frac{i}{2} \Gamma)^2 \). Since \( \Omega \propto \sqrt{I} \), we can substitute in (4.3) \( \Omega^2 = r I \), where \( r \) is a parameter to be determined from the fit. We fit positions of resonances \( a, b \) with function \( f_{01} \) and \( c \) with \( f_{12} \), where “01” and “12” indices stand for the \( |m_g = 0\rangle \leftrightarrow |m_g = \pm 1\rangle \) and \( |m_g = \pm 1\rangle \leftrightarrow |m_g = \pm 2\rangle \) transitions, respectively. Obviously, if our model is correct, all fits should produce the same value of the \( r \) parameter.

Results of the fitting procedure for both probe polarizations are shown in Figure 4.10. For the \( \sigma^- \)-polarized probe we obtain almost equal value of the scaling factor \( r = 0.38 \pm 0.01 \) for resonances \( a, b \) and \( c \). In the case of \( \sigma^+ \)-polarized probe, however, the fit with \( r = 0.38 \) results in a large error for resonances \( a \) and \( b \). The value of \( r \) that fits best these resonances is 0.44. Moreover, for resonances \( a \) and \( b \), there is a systematic deviation of experimental points from function \( f_{01} \) (4.3). This deviation is bigger in the case of \( \sigma^+ \)-polarized probe and is even more evident that disagreement of \( r \)

![Figure 4.10: Positions of resonances in the ABS spectra plotted as a function of intensity of the trapping beams (I is the intensity per beam) and fitted with function (4.3) for the \( \sigma^+ \) and \( \sigma^- \) probe polarization. For better evaluation of its symmetry with \( b \), the sign of position of \( a \) was changed.]
values in the discussed fits. It suggests that either \(a\) and \(b\) cannot be associated with the discussed RLSS processes or there are some other resonant contributions that influence position of \(a\). Another argument that supports the latter conjecture is the ultra-narrow resonant structure that appears in the \(\sigma^-\) ABS spectra and is marked in Figure 4.8.

Moreover, the FWM spectra for both probe beam polarizations also exhibit ultra-narrow structures in the very vicinity of \(\delta = 0\). In contrast to the ABS spectra, these structures are not overwhelmed by other resonances. It shows that FWM spectroscopy is far more sensitive than ABS spectroscopy. The value of \(r\) allows one to determine the average Rabi frequency \(\Omega\) in (4.3) which for \(I = 95\) \(\mu\)W/mm\(^2\) per beam amounts \(6\Gamma\).

We can model CS in the ABS spectra with expression (1.94) (page 36). However, rather than plotting (1.94) for one value of the Rabi frequency, we shall use a certain range of \(\Omega\) values, like we did in Section 4.3. This modelling is shown in Figure 4.11. Inset in this Figure depicts the spectrum generated for \(\Omega = 6\Gamma\) with the resolved resonances prior to averaging.

Although the shape of the experimental signal is qualitatively well reproduced, some of its features cannot be explained. Firstly, the slope of the experimental spectrum around \(\delta = 0\) is steeper than in our simulation. Secondly, the model discussed above fails to explain the differences between the spectra recorded for \(\sigma^+\) and \(\sigma^-\)-polarized probe. These drawbacks of our model call for inclusion of additional mechanisms for explanation of ABS and FWM lineshapes.

### 4.4.3 Contribution of RIR to the spectra

In the previous section we found that the central part of the ABS and FWM spectra is influenced by processes other than RLSS. Additionally, our attention was drawn to the ultra-narrow structure in the very vicinity of \(\delta = 0\) of the ABS spectra recorded for the \(\sigma^-\)-polarized probe beam, i.e. when the probe and the nearest trapping beam have the same polarizations. To approach well-determined conditions of a single pump beam pair, we performed a series of measurements in which we increased the power in the pair that was nearly collinear with the probe at the expense of the two other pairs, as shown in Figure 4.12.

By creating such an intensity imbalance we approach a 1D configuration of pump-probe spectroscopy (in the limiting cases almost whole power in sent into the pair of trapping beams closest to the probe). Due to the increase of radiation pressure along the \(z\) axis, the cloud of cold atoms is being compressed in this direction so the magnetic field effect, discussed in the forthcoming section, becomes smaller. The polarization configuration of the net trapping field is also altered – the dominant polarization contribution is brought due to the trapping beams pair nearly co-linear with the probe. This is the previously discussed \(\sigma^+ - \sigma^-\) configuration of the counter-propagating beams (Section 2.5, page 48).

From the experimental point of view, the possibility of maintaining a MOT for the imbalanced intensity between pairs of trapping beams requires their good geometrical adjustment and accurate compensation of any residual magnetic field in the trap (see Section 3.4, page 62). Fulfilling these demands resulted in a perfect stability of the atomic cloud also for the most uneven power division between the trapping beams, when almost all light was sent to \(z\) pair.

The ABS and FWM spectra are presented in Figures 4.13 and 4.14. When com-
Figure 4.11: Modelling of the experimental spectra obtained for $I = 102 \, \mu W/mm^2$ and $\sigma^-$-polarized probe beam. The range of Rabi frequencies used for spectra averaging is $[4.8\Gamma; 7.8\Gamma]$. The width of the Lorentz functions in formula (1.94) is $0.13\Gamma$ and corresponds to the ground level broadening due to admixture of the excited level (in the dressed-atom model). Inset shows the level scheme and the resolved contributions plotted for specific value of $\Omega = 6\Gamma$. The resonances in the inset are labelled by the $m_g$ numbers of the sublevels involved in corresponding transitions.

pared with Figures 4.8 and 4.9 for equal trapping beam intensities, these spectra reveal very interesting features. Firstly, in Figure 4.13 the ultra-narrow resonant structure for $\sigma^-$ evolves into a dispersive resonance whose shape, width and sign agree with RIR. Secondly, the $\sigma^+$ spectra, which have a smooth appearance for low $I_z$, exhibits a distinct steep-sloped structure for higher $I_z$. As can be seen, the differences between $\sigma^+$ and $\sigma^-$-polarized probe spectra become more pronounced for higher intensity imbalance.

To explain the central part of the CS in the ABS spectra we shall keep in mind that the contribution of recoil effects (RIR) in the close vicinity of $\delta = 0$ results from the Raman transitions with $\Delta m_g = 0$. Thus, the pump and probe photons involved in such transitions must have the same polarization. This has significant consequences for the relative amplitudes of the contributions to the RIRs, associated with various trapping beams.

For example, if the probe is $\sigma^-$ polarized, only the trapping beams that generate the net $\sigma^-$ polarization contribute to the RIRs. For small $\theta$, this condition immediately selects the nearly co-propagating trapping beam, i.e. the one which makes a small angle $\theta$ with the probe. Similarly, for the $\sigma^+$ polarization only the counter-propagating beam contributes to the signal. In addition to these collinear
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Figure 4.12: Increase of power in the pair of trapping beams propagating almost co-linear with the probe beam at the expense of two other pairs as the approximation of 1D pump-probe configuration. The 1D situation is best approximated for the most uneven power division $I_z \gg I_x, I_y$, where $I_i$ stands for the intensity of the beam propagating along $i$ axis. In the case of our measurements, we have always $I_x = I_y$.

beams, there is also a contribution of the four transverse beams (precisely speaking, making angles $90^\circ$ and $90^\circ \pm \theta$ with the probe). These transverse beams, viewed along the axis of the probe (approximately $z$), appear linearly polarized in the $x$ and $y$ directions. Their superposition creates an electric field of arbitrary polarization, depending on the relative phases. In particular, the transverse beams generate the $\sigma^+$ field with the same probability as the $\sigma^-$ field. For this reason, they make equal contributions to the RIR seen with $\sigma^+$ and $\sigma^-$ probe polarizations. Consequently, the main difference between the ABS spectra for two opposite polarizations of the probe is caused by the different widths of RIR for $\theta$ and $180^\circ - \theta$ pump-probe orientations.

We can verify whether our assumption that the steep resonance in the center of the $\sigma^+$ ABS spectra is due mainly to the RIR for $180^\circ - \theta$ pump-probe angle is correct. For small $\theta$, the ratio of the RIR resonance widths for $\theta$ and $180^\circ - \theta$ is simply

$$\frac{2\delta_{\text{res}}(\theta)}{2\delta_{\text{res}}(180^\circ - \theta)} \approx \frac{2k_u\theta}{4k_u} = \frac{\theta}{2}.$$  \hspace{1cm} (4.4)

By determining the widths of the corresponding resonances we obtain the value of $\bar{\theta} = 3.4^\circ \pm 0.4^\circ$. The average $\bar{\theta}$ value results from determination of $\theta$ from all pairs of the $\sigma^+$ and $\sigma^-$ ABS spectra. It agrees quite well with the pump-probe angle measured in our experiment ($3.3^\circ \pm 0.2^\circ$). The example of determination of $\theta$ for particular pair of $\sigma^+$ and $\sigma^-$ ABS spectra is shown in Figure 4.15. From the width of resonances we obtain $\theta = 3.3^\circ$ and $T = 76 \mu$K, which is consistent with the TOF temperature measurements [98]. However, there are some limitations in precision of determination of $\theta$ and $T$ from the experimental ABS signals. These limitations are due to the approach used to extract the RIR widths from the ABS spectra. This
Figure 4.13: Series of the ABS spectra recorded for $\sigma^+$ and $\sigma^-$-polarized probe beams. $I_z$, the intensity in the pair of the trapping beams, (indicated beside every plot) has been increased at the expense of the other pairs, as shown in Figure 4.12. When all trapping beams are equally strong, the intensity per one beam equals 120 $\mu$W/mm$^2$. Trap beams detuning $\Delta = -3\Gamma$, axial magnetic field gradient $\frac{\partial B}{\partial z} = 13$ Gauss/cm.
Figure 4.14: Series of the FWM spectra. Experimental conditions are the same as in the previous Figure.
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Approach can be summarized as follows:

1. The RIR contribution for the angle $180^\circ - \theta$ is not well distinguished from the RLSS structure, since the RIR amplitude and width are comparable to those of the $a$ and $b$ RLSS resonances. The width of the $180^\circ - \theta$ RIR can be determined by a fit of the Gaussian derivative to the slope around $\delta = 0$, as shown in Figure 4.15. Verification of the fit agreement with the experimental spectrum is additionally supported by the “kinks” in the ABS spectrum (marked in Figure 4.15 by short, block arrows), the evident signature of additional resonant contributions different from RLSS. Moreover, the central part of the ABS spectra, in addition to the RLSS contribution, is influenced by RIR resonances due to the trapping beams making $90^\circ$ angle with probe, which is another source of fit inaccuracy.

2. For small angle $\theta$, the RIR resonances are very narrow with their widths approaching the resolution limit of the experiment. Thus, the uncertainty of their

Figure 4.15: Verification whether the central part of CS in absorption spectra is influenced by the RIR contributions. By determining the ratio of width of the RIR for $180^\circ - \theta$ pump-probe angle ($\sigma^+$ spectrum) to the width of the RIR for small pump-probe angle $\theta$ ($\sigma^-$ spectrum), we find $\theta$ value ($3.3^\circ$ for the presented case). Since during the experiment $\theta$ was not changed, the ratio should produce the same value independent of experimental conditions. We performed such a test for all $\sigma^+/\sigma^-$ pairs of the spectra with sufficiently resolved RIR contributions and obtained the average value $\bar{\theta} = 3.4^\circ \pm 0.4^\circ$ ($\theta$ measured in experiment is $3.2^\circ \pm 0.2^\circ$). The spectra presented in this Figure are recorded for $I_z = 300 \mu W/mm^2$. Short block arrows show the “kinks” discussed in the text.
width determination become bigger. Moreover, like in the $180 - \theta^\circ$ case, the small-angle RIRs also appear on the background of RLSS and $90^\circ$ RIR.

Despite the drawbacks listed above, we have obtained a good agreement with the actual experimental situation in terms of angle $\theta$ and temperature of the cloud. This supports conclusion that central part of the CS is substantially affected by recoil-induced resonances.

### 4.4.4 Modelling of spectra with recoil-induced resonances

Having verified that the central part of CS is influenced by RIRs, we can model the spectra including the RIR contributions. Let us denote the RLSS contribution, given by the equation (1.94), page 45, as $s_{RLSS}(\delta)$ and the RIR contribution, given by equation (2.14), page 45, as $s_{RIR}(\delta, \theta)$. The ABS spectrum for a single Rabi frequency $\Omega$ will then be modelled by equation

$$s(\delta) = s_{RLSS}(\delta) + \sum_{\sigma^\pm; 90^\circ - \theta, 90^\circ + \theta} s_{RIR}(\delta, \theta),$$

(4.5)

where summation of the RIR contributions over angles refers to the relevant polarizations of the probe beam, as discussed in the previous Section. Expression (4.5) should be averaged over a certain range of Rabi frequencies, like we did in Figure 4.11. We shall keep in mind, however, that only the RLSS contribution is sensitive to this kind of averaging.

![Diagram](image)

Figure 4.16: Experimental ABS spectra for $\sigma^-$ and $\sigma^+$-polarized probe beam modelled according to equation (4.5). Experimental conditions and RLSS averaging parameters are the same as in Figure 4.11. Temperature of the cloud was chosen $T = 130 \ \mu K$ for the best agreement of the simulation with experimental data.
The result of simulation of equation (4.5) corresponding to the same experimental spectra as in Figure 4.11 is shown in Figure 4.16. The agreement of the simulated shape of CS is obviously better now than in the case of Figure 4.11, where the RIR contribution was not included. Inclusion of the RIR allows one not only to obtain a good agreement with the experimental data for the slope in the central part of CS, but also to explain the qualitative differences between the ABS spectra recorded for the $\sigma^+$ and $\sigma^-$-polarized probe beams. We are able to reproduce the relative amplitudes between the $b$ and $c$ resonances and understand the origin of their different widths. Also, the amplitude of $a$ over the rising pedestal for $\delta < 0$, different for the $\sigma^+$ and $\sigma^-$-polarized probes, is well reproduced by our model. Finally, theoretical ABS spectra obtained with our present model possess the ultranarrow structure $R$ observed in the ABS spectra for the $\sigma^-$ probe polarization. The drawback of the model is the insufficient width of the resonances, which is seen especially in the $c$ resonance. This is the reason why the $b$ and $c$ resonances are resolved in the $\sigma^+$ ABS spectrum in Figure 4.16, in contrast to the relevant experimental signal. This broadening will be discussed in the next section.

### 4.4.5 Influence of quadrupole magnetic field on the spectra

Atoms in a MOT gather in a point where the quadrupole magnetic field is zero. Since the size of the cloud is finite, atoms whose coordinates differ from zero are under the influence of a non-zero magnetic field $\mathbf{B} \neq 0$. Sublevels of the atomic ground state are thereby Zeeman-shifted depending on the atom position in the trap. Thus, the positions of Raman resonances involving light-shifted ground states will be altered by $\mathbf{B}$ (Figure 4.17). This causes an inhomogeneously broadening, which can be responsible for the widths of resonances discussed in the previous sections.

![Figure 4.17: Gaussian spatial distribution of atoms in a MOT, $p(z)$ ($\sigma_z = 1$ mm) and frequency shift $\Delta \nu$ of the RLSS due to quadrupole magnetic field $\mathbf{B}$ (shown for $\frac{\partial B}{\partial z} = 13$ Gauss/cm).](image)

Let us consider the one-dimensional situation. The probability $p(z)\,dz$ of finding
an atom along between $z$ and $z + dz$ is given by the Gaussian distribution

$$p(z) dz = \frac{1}{\sqrt{2\pi} \sigma_z} \exp \left[ -\frac{z^2}{2\sigma_z^2} \right] dz,$$  \hspace{1cm} (4.6)$$

where $\sigma_z$ is the Gaussian radius of a cloud. The RLSS processes occurs between adjacent sublevels of the ground state ($\Delta m_g = \pm 1$). Hence, introducing $\delta_{\text{res}}$ as a resonant pump-probe detuning for an atom at $z = 0$, we obtain the Raman resonant condition for an atom at $z$ in the form

$$\delta(z) = \delta_{\text{res}} + \Delta m_g \frac{g_F \mu_B}{\hbar} \frac{\partial B}{\partial z} z,$$  \hspace{1cm} (4.7)$$

where $g_F$ is the Landé factor associated with the $^2S_{1/2}(F = 3)$ state and $\mu_B$ is Bohr magneton. Since the resonance amplitude is proportional to the number of atoms fulfilling the resonant condition (4.7), by elimination of $z$ from (4.7) and substituting it into (4.6), we obtain the lineshape of an inhomogeneous broadened resonance

$$s(\delta) = C \cdot \exp \left[ -\frac{(\delta - \delta_{\text{res}})^2}{2\sigma_B^2} \right],$$  \hspace{1cm} (4.8)$$

where

$$\sigma_B = \frac{g_F \mu_B}{\hbar} \frac{\partial B}{\partial z} \sigma_z,$$  \hspace{1cm} (4.9)$$

Figure 4.18: Comparison of the experimental ABS spectra with theoretical simulations, which include inhomogeneous broadening due to magnetic field $B$ generated by quadrupole trapping coils. Experimental conditions: trapping beams intensity $I = 102 \mu W/mm^2$ per beam, detuning $\Delta = -3\Gamma$, axial magnetic field gradient $\frac{\partial B}{\partial z} = 13$ Gauss/cm. Parameters of theoretical simulation: Rabi frequency averaging range $\Omega \in [5\Gamma; 7.5\Gamma]$, Gaussian width of a single, inhomogeneously broadened resonance $\sigma_B = 0.088\Gamma$, which corresponds to the trap size $\sigma_z = 0.9$ mm, temperature $T = 155 \mu K$ was chosen for the best agreement of the simulation with experimental data.
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Figure 4.19: Experimental ABS spectra for $\sigma^+$ and $\sigma^-$ polarized probe beams for various magnetic gradients (noted beside each plot). Decrease in $\frac{\partial B}{\partial z}$ results, as expected, in better resolved individual RLSS resonances. The broadening of the spectra for big values of $\frac{\partial B}{\partial z}$ can be additionally caused by high collision ratio in compressed trap. Experimental parameters: trapping beam intensity $I = 150 \mu W/mm^2$, trapping beams detuning $\Delta = -2.9\Gamma$. 
and $C$ is the resonance amplitude. The derivation was performed with assumption that the width of resonance curve [defined by (1.95), page 37] $\gamma_{i,i+1} \ll \sigma_B$. Due to the inhomogeneous broadening caused by the quadrupole magnetic field we can simulate the experimental spectra replacing the Lorentz functions in RLSS part of (4.5) by Gaussians. The result of such modelling is shown in Figure 4.18. Comparison of 4.16 and 4.18 shows importance of the inhomogeneous broadening in explanation of the experimental data.

To illustrate better this importance, we have performed a series of measurements with different $B$ gradient. The results are shown in Figure 4.19. Indeed, the resonances become better resolved for smaller values of $\frac{\partial B}{\partial z}$. We should keep in mind, however, that for smaller magnetic field gradient the size of the cloud increases. Thus, the broadening reduces due to the decrease of $\frac{\partial B}{\partial z}$, but increases due to the $\sigma_z$ increase [see equation (4.9)]. However, in Ref. [83] it is shown that $\sigma_z \propto I_{\text{coil}}^{-1/2}$, where $I_{\text{coil}}$ is the current that flows in the quadrupole coils. Because $\frac{\partial B}{\partial z} \propto I_{\text{coil}}$, the growth of $\sigma_z$ does not fully compensate for the decrease of $\frac{\partial B}{\partial z}$, we observe the net narrowing of the RLSS resonances and thereby better resolved spectra. A broadening of the spectra for big values of $\frac{\partial B}{\partial z}$ can be additionally caused by high collision ratio in a compressed trap.

Another evidence of the important role of the magnetic field broadening is provided by the dependence of the ABS spectra on power in the trapping beam pair along $z$. When this power is increased (Figure 4.13), the spectra exhibit narrowing of the RLSS contribution, although not as pronounced as in Figure 4.19. This effect is due to compression of the atomic cloud along $z$, resulting from the increased radiation force in this direction. These observations clearly demonstrate important role of inhomogeneous broadening in the cold atom spectra. To author’s knowledge, such broadening has never been discussed before.

### 4.4.6 Remarks on quality of the ABS and FWM spectra

Inclusion of the magnetic field effects in the description of the processes that govern the CS not only allows us to explain its shape, but also helps to diagnose experimental properties of our setup. We noticed that the signal quality strongly depends on the precision of MOT adjustment. We observed that the best signals are obtained when the cloud is spherically symmetric and expands isotropically after switching off the quadrupole magnetic field. Such a behavior is intrinsically related to perfect compensation of a residual magnetic field (including the Earth magnetic field) and to precise alignment of the geometry and power balance in the trapping beams. These factors guarantee that atoms gather in the point where the magnetic field is indeed zero so that the effect of its gradient is minimized. If the cloud is being pushed out of this optimal position, the resolution of the spectra drops and the symmetry of the relevant resonance positions relative $\delta = 0$ is perturbed.

Thus, before each set of measurements was performed, we checked the parameters of the cloud in terms of its optimal adjustment. By tweaking of the mirrors that guide trapping beams and/or by changing the current in compensation coils we were able to shift the cloud about 1 mm, i.e. by about its size. Such shift can substantially modify positions of the RLSS resonances and blur the spectra (see Figure 4.17 for more qualitative picture of the situation). Very careful adjustments of the trap and fortunate combination of its experimental parameters allowed us to register well-resolved ABS spectra, like the one displayed in Figure 4.20, where many resonant
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Figure 4.20: Experimental ABS spectra for $\sigma^+$ and $\sigma^-$ polarized probe beams. Almost whole power of the trapping light is in the pair of beams closest to the probe (intensity $I_z = 310 \mu\text{W/mm}^2$ per beam, intensity of the remaining trapping beams $I = 10 \mu\text{W/mm}^2$ per beam), detuning of the trapping beams $\Delta = -4\Gamma$. The presented ABS spectra reveal many resonant contributions (pointed by arrows), which refer to the discussed RLSS processes. In addition, a very weak, yet visible resonance appears (marked by asterisk) that is beyond our model and possibly connected with the RLSS for other than $\pi - \sigma^\pm$ pump-probe polarization configurations. In these spectra, the RIR structures are very well resolved and are marked by the double arrows.

contributions are discernible. Unfortunately, such adjustment was so critical and unstable that no systematic study of the ABS and FWM spectra could be performed (too small or too unstable clouds, with too few atoms to provide measurable FWM signal generation).

### 4.4.7 Rayleigh resonance in the ABS spectra

In Ref. [53] Raman spectroscopy was used to determine the friction coefficient connected with the atomic motion in a 1D corkscrew optical molasses, i.e. in the light field created by $\sigma^+ - \sigma^-$ counter-propagating laser beams. Lounis et al. [53] first prepared atoms in a MOT, then switched-off the trapping fields (optical and magnetic) and switched-on independent $\sigma^+ - \sigma^-$ molasses beams together with a circularly polarized probe beam making a small angle $\theta$ with one of the molasses beams. In the case when the polarization of the probe was the same as the co-propagating molasses beam, a narrow dispersion-like resonance appeared on the absorption spectra close to $\delta = 0$. The peak-to-peak width of this resonance equals $2\beta/m$, where $m$ is the mass of an atom and $\beta$ is the friction coefficient that the moving atom experiences when travelling in the molasses\(^3\).

We were able to observe the Rayleigh resonance in a working MOT, even in the case when all the trapping beams were power-balanced. The Rayleigh resonance appears in the ABS spectrum for small trapping beam detuning ($\Delta = -\Gamma$ and $\Delta = -2\Gamma$) and $\sigma^-$-polarized probe beam. It exhibits the same behavior as reported in Ref. [53]. For $\Delta \geq -3\Gamma$ and more, the Rayleigh resonance is eventually replaced.

\(^3\)The radiation trapping force acting on atom is $F = -\beta v$, $v$ is atomic velocity [52].
Figure 4.21: Series of the ABS spectra measured for the trapping beam detuning $\Delta = -\Gamma$, $-2\Gamma$ and $\sigma^-$-polarized probe beam. The intensity in the trapping beams pair closest to the probe beams was set to the value shown at each plot. The intensity per one beam in the case of equal power division between all trapping beams was $I = 120 \mu W/mm^2$, $\frac{\partial I}{\partial z} = 13$ Gauss/cm. The spectra exhibit dispersive Rayleigh resonance at $\delta = 0$ which is the sign of sub-Doppler cooling in the corkscrew molasses [53]. For higher intensities, the Rayleigh resonance becomes split and for larger detuning $\Delta$ it is eventually replaced by RIR.
4.5 Conclusions and outlook

We have performed a series of measurements that fully document spectroscopic properties of a working MOT with all optical and magnetic fields turned on. We recorded the ABS and FWM signals simultaneously, hence no systematic errors appear in the direct comparison of the spectra and their interpretation, yielding complementary information. The spectroscopy of our MOT, performed in wide and narrow scan regimes, gave us valuable information about the light-atom interaction in the trap.

We have found that while the wide-scan spectra can be qualitatively understood in terms of the dressed atom model, the full quantitative explanation of their shapes requires the DTLS theory and inclusion of the light intensity and polarization modulation in the trap.

We used the numerical code developed by Aurél Gábris [44], which takes into account the DTLS theory for any angular momenta values. The code has been tested for various configurations of the probe-pump polarization in qualitative and quantitative aspect (Section 1.13 and 1.14, page 27). Only in the case of the central structure in the $\pi - \sigma^+$ configuration the spectra obtained with the program and simple Raman transition model (1.94, page 36) do not exactly agree. The reasons of
failure of the code in this particular case will be carefully examined. Since it was the only case when the code gave suspicious results, its output has been successfully used to model the ABS and FWM wide-scan spectra and to explain the shift between the ABS and FWM sideband positions (4.3).

The narrow-scan spectra which were used to study the central structure (CS) are certainly more interesting than the ones recorded in the wide-scan regime. Contributions to the CS result from Raman transitions between energy levels in structures of two kinds:

1. Energy levels of an atom, altered by the net trapping field - the so-called “internal degrees of freedom”.

2. Kinetic energy levels, either discrete, connected with the atomic localization and optical lattices, or continuous, associated with a free motion of the unbound atoms confined in a trap - the, so-called, “external degrees of freedom”.

Raman spectroscopy can be used not only to investigate light-atom interaction in terms of atomic energy levels, but also to determine the mechanical and statistical properties of the cold atomic sample, confined in a trap and subject to the intense field of several trapping beams. The analysis of the recorded ABS spectra performed in this chapter resulted in identification of the following processes that govern the shape of the CS:

1. RLSS – Raman transitions between light-shifted sublevels. The theoretical treatment used to explain the shape of the RLSS contribution to the spectra was performed in the for $\pi - \sigma^\pm$ pump-probe polarization configuration introduced in the Section 1.14.2, page 36.

2. RIR – recoil-induced resonances. Inclusion of the atomic recoils in the net field generated by all trapping beams was necessary to understand the spectra in the very vicinity of $\delta = 0$ and their dependence on the probe polarization.

3. Rayleigh resonance - the spectroscopic manifestation of sub-Doppler cooling in the $\sigma^+ - \sigma^-$ corkscrew molasses created by the pair of trapping beams nearly co-linear with the probe.

We have excluded from our considerations the Raman processes associated with optical lattices (RVOL). This is due to instability of the relative phases of the trapping beams that precludes atomic localization and the lack of clear signature of the typical spectroscopic features associated with the RVOL processes in the recorded spectra.

To obtain satisfactory agreement between our modelling and experimental spectra, the following processes that are responsible for broadening and shaping of the signals have been taken into account:

1. Modulation of a net field intensity in the trap, and consequently of the Rabi frequency. Averaging theoretical spectra over the range of Rabi frequencies produced far better agreement than the assumption of its stable value (Figure 4.11).
2. Inhomogeneous broadening caused by influence of the quadrupole magnetic field on the sample. Inclusion of this phenomenon resulted in the theoretical ABS spectra that almost perfectly reproduced the experimental signals (Figure 4.18). To the best of author’s knowledge this aspect of MOT magnetic field has never been discussed before.

FWM spectra, however, are not yet quite understood. The complicated shape of the FWM spectra and their evident dependence on the probe beam polarization, even more pronounced than in the case of the ABS spectra, originates from interference of the terms connected with the RLSS/RIR/RVOL and Rayleigh contributions. Consequently, theoretical modelling of the FWM spectra is more difficult than the ABS signals. Moreover, as it was discussed in the case of the wide-scan spectra, some pump-probe polarization configurations that modify ABS spectra do not participate in four-wave mixing. This fact is consistent with our observation that the positions of CS resonances in the FWM spectra do not correspond with positions of resonances in the ABS signals. The FWM spectra will be subject of further studies. As it can be seen from the FWM spectra presented in this thesis, the FWM spectroscopy constitutes a very promising tool for probing the mechanical properties of the cold atomic sample. Its sensitivity and the resolution of ultra-narrow structures that originates from atomic motion is much better than in case of ABS spectroscopy. Still, further work, mainly on theoretical basis is needed to fully exploit all these potentials.
Appendix A

Light shifts of unstable states

In this appendix we derive expression for the light shift and width of dressed atomic levels in the limit of small ($\Omega \ll |\Delta|, \Gamma_e, \Gamma_g$) and high ($\Omega \gg \Gamma_e, \Gamma_g$) laser beam intensities ($\Gamma_g$ and $\Gamma_e$ are the decay rates of ground and excited states, respectively). In the following we use tilde to distinguish between complex (with tilde) and real variables.

The derivation generalizes the problem from Section 1.3 by inclusion of finite lifetimes of the atomic levels and is based on diagonalization of the Hamiltonian (see Figure A.1)

$$\hat{H} = \hbar \left( \omega - i \frac{\Gamma_g}{2}, \frac{-i}{2} \Omega - \frac{i}{2} \Omega_0 - \frac{i}{2} \Gamma_e \right).$$

The eigenvalues of $\hat{H}$ are given by equation analogous to (1.37)

$$\tilde{\varepsilon}_\pm = \frac{\hbar}{2} \left( \tilde{\omega} + \tilde{\omega}_0 \pm \tilde{\Omega}' \right),$$

where

$$\tilde{\omega} = \omega + i \frac{\Gamma_g}{2},$$

$$\tilde{\omega}_0 = \omega_0 + i \frac{\Gamma_e}{2},$$

$$\tilde{\Omega}' = \sqrt{\Omega^2 + \tilde{\Delta}^2},$$

$$\tilde{\Delta} = \Delta + i \frac{1}{2} (\Gamma_e - \Gamma_g).$$

Figure A.1: Two-level atom unstable states. The right part of the picture shows the atom-field states in the $|i\rangle \otimes |N\rangle$ basis (see page 10) and explains the form of Hamiltonian (A.1).
Let us consider $\tilde{\Omega}'$ in the limit $\Omega \ll |\Delta|, \Gamma_e, \Gamma_g$. First, in order to take care of the sign of $\Delta$ we ignore the imaginary part of $\tilde{\Delta}$ in (A.6). Expansion of $\tilde{\Omega}'$ yields

$$\tilde{\Omega}' \approx \begin{cases} -\Delta - \frac{1}{2} \Omega'^2 / \Delta, & \Delta < 0 \\ \Delta + \frac{1}{2} \Omega'^2 / \Delta, & \Delta > 0. \end{cases} \quad (A.7)$$

By replacing $\Delta$ with $\tilde{\Delta}$ and separating the real and imaginary parts, we get

$$\tilde{\Omega}' \approx \begin{cases} -\Delta(1 + s) - i \frac{\Gamma_e - \Gamma_g}{2}(1 - s), & \Delta < 0 \\ \Delta(1 + s) + i \frac{\Gamma_e - \Gamma_g}{2}(1 - s), & \Delta > 0. \end{cases} \quad (A.8)$$

where we introduced the saturation parameter $s$

$$s = \frac{\Omega'^2/2}{\Delta^2 + (\Gamma_e - \Gamma_g)^2/4}. \quad (A.9)$$

Note that for $\Gamma_e = \Gamma$ and $\Gamma_g = 0$, $s$ takes the form used in equation (2.31). Substituting (A.8) in (A.2), we obtain expressions for the energy of levels $|g\rangle$ and $|e\rangle$, modified by interaction with light:

$$\tilde{\varepsilon}_{LS,|g\rangle} = \begin{cases} \tilde{\varepsilon}_-, & \Delta < 0 \\ \tilde{\varepsilon}_+, & \Delta > 0 \end{cases} \approx \hbar \left( \omega + \frac{\Delta}{2}s \right) - \frac{i}{4} \left( \Gamma_e - \Gamma_g \right) \left( \Gamma_e - \Gamma_g \right) \frac{\Delta}{\Omega'} \quad (A.10)$$

$$\tilde{\varepsilon}_{LS,|e\rangle} = \begin{cases} \tilde{\varepsilon}_+, & \Delta < 0 \\ \tilde{\varepsilon}_-, & \Delta > 0 \end{cases} \approx \hbar \left( \omega_0 - \frac{\Delta}{2}s \right) - \frac{i}{4} \left( \Gamma_e - \Gamma_g \right) \frac{\Delta}{\Omega'} \quad (A.11)$$

From equations (A.10,A.11) and for $\Gamma_g < \Gamma_e$ one concludes that the near-resonance state $|g\rangle$ broadens and $|e\rangle$ becomes narrower, as it is shown in Figure 1.4b.

In the case of high laser beam intensity, $\Omega \gg \Gamma_g, \Gamma_e$, the generalized complex Rabi frequency can be written as

$$\tilde{\Omega}' \approx \Omega' + i \frac{\Gamma_e - \Gamma_g}{\Omega'} \frac{\Delta}{2}. \quad (A.12)$$

In the expansion (A.12), term of the order of $(\Gamma / \Omega')^2$ was dropped ($\Omega' = \sqrt{\Omega'^2 + \Delta^2}$). According to (A.2), the energies of $|\pm\rangle$ dressed levels can be approximated as

$$\tilde{\varepsilon}_{\pm} \approx \hbar \left( \omega_0 + \frac{\Delta}{2} \pm \frac{\Omega'}{2} \right) - \frac{i}{4} \left( \Gamma_e + \Gamma_g \mp \frac{\Delta}{\Omega'} (\Gamma_e - \Gamma_g) \right). \quad (A.13)$$

It is easy to verify that for $\Delta = 0$, both dressed levels have the same width, $\frac{1}{4}(\Gamma_e + \Gamma_g)$. For $\Delta \to \infty$, the width of level $|+\rangle$ tends to $\Gamma_g$, whilst the width of level $|-\rangle$ approaches $\Gamma_e$. This conclusions are reflected in the plot depicted in Figure 1.4a.
Appendix B

Calibration of the frequency scale

As it has been discussed in Chapter 3, the probe laser is tuned indirectly by driving it with the injected master laser beam after double pass through the acousto-optic modulator (AOM-1) (Figure 3.2, Section 3.3). As mentioned in Section 3.3, the AOM-1 is frequency swept using a linear voltage ramp connected to the input of a VCO-based controller. By capturing this voltage simultaneously with the ABS and FWM spectra, we can assign relevant voltage to each point of the frequency axis in the recorded spectra. Having already measured voltage-frequency characteristics of all VCOs in the experiment\(^1\), we can calculate detuning \(\delta\) according to the formula derived in Section 3.3, namely

\[
\frac{\delta}{2\pi} = f_{AOM-2} + f_{AOM-3} - 2f_{AOM-1},
\]

where \(f_{AOM-i}\) is the frequency of AOM-i.

Theoretically, to get accurate value of the detuning, it is sufficient to know the voltage levels on all VCOs that drive AOMs and to use them for frequency evaluation by (B.1). In practice, however, when we measure detuning in that way systematic errors connected with the determination of three AOM frequencies shift the frequency axis. Since our spectra are studied mainly in the very vicinity of \(\delta = 0\) and we have to check whether the ultranarrow structures that appear in the recorded spectra are indeed centered at \(\delta = 0\), it is essential to eliminate such uncertainty. Experimental identification of the recoil-induced and Rayleigh resonances is greatly supported by precise determination of the \(\delta = 0\) point on the frequency axis.

For that purpose we superimposed the probe beam with one of the trapping beams on the detector and observed the beatnote produced by their interference as the function of voltage applied to the VCO driving AOM-1. In the case of perfect matching of the probe and trapping beam frequencies, \(\delta = 0\), the beatnote should approach a DC signal. For small departures of \(\delta\) from zero, the beatnote exhibits sinusoidal modulation in a kHz range. The result of this experiment is shown in Figure B.1 in the form of oscilloscope screen frames for various voltage levels around the value for which \(\delta = 0\). The minimum beatnote frequency that we were able to measure was about 3 kHz. It was impossible to obtain the DC beat signal due to finite precision of the voltage sources used in experiment\(^2\). We also observed

\(^1\)It was done using precisely controlled DC voltage source from Stanford Research SR830 DSP Lock-In Amplifier and frequency counter Dagatron 7023.

\(^2\)Minimal step of the digital voltage source used in this experiment (1 mV) was too big to reach better precision.
a FM modulation of the beatnote signal which is due to short-term fluctuations of the VCOs and additionally limits our resolution. These measurements proved that relative frequency of the lasers can be controlled with precision of at least 3 kHz. Moreover, we have checked that the voltage corresponding to the minimum beatnote frequency coincides with the center of ultra-narrow resonances observed in the ABS and FWM spectra, which is the most important conclusion for analysis of the spectra.

Figure B.1: Beatnote of the probe and one of the trapping beams registered for various voltage levels driving the VCO responsible for the probe beam tuning. The minimum beatnote frequency (about 3 kHz) was registered for 93 mV. This voltage coincides with the ramp level for which the center of ultra-narrow structures in the ABS and FWM spectra is observed. Time scale in the oscilloscope screen frames is 100 $\mu$s/div.
Appendix C

Stability of the trapping beam phases

In the case of $\sigma^+ - \sigma^-$ MOT beams configuration, the net optical field depends significantly on the relative phases of the beams. In contrast to the case of $N+1$ beams creating $N$-dimensional lattice [79], any change of the relative phases of MOT beams not only shifts the modulation pattern as a whole, but also alters its topology. Consequently, various configurations of the trapping beam phases lead to different realizations of optical lattice. Thus, in order to obtain atomic localization in a MOT, one has to control the trapping beam phases, like it was done in [81]. Fast, random fluctuations of the MOT beam phases precludes stable atomic localization. However, if these fluctuations exhibited sufficiently long periods of phase stability, one could expect some localization, which could have impact on the probe-beam spectra.

![Figure C.1: (a) Experimental setup for measuring relative phases of the MOT beams. Interference pattern created by the $x$ and $z$ beams is monitored by a small photodiode (dark spot in the interference pattern). (b) Measured phase fluctuations for two timescales. Plots on the right show a huge phase-noise generated by vibrations of a fan in air-purification system.](image)

In order to test the phase stability of our setup, we have performed a simple experiment. We measured temporal dependence of the interference pattern produced by superposition of the trapping beams propagating in the $x$ and $z$ directions (Figure C.1a). Small photodiode BPY P42 monitored the shift of interference fringes. To
superimpose the $x$ and $z$ beams it was necessary to change orientation of the $\frac{\lambda}{4}$ waveplates by retroreflection mirrors in the MOT arms. The results, displayed in Figure C.1b, show that the phases exhibit fast, random fluctuations which are very sensitive to mechanical vibrations in the lab. This test demonstrates that stable optical lattices in our MOT are very unlikely.
Appendix D

Interference pattern of the MOT beams

Three pairs of counter-propagating beams in the \( \sigma^+ - \sigma^- \) configuration intersect in the trapping region and create spatial intensity and polarization modulation of the net light. Below, we calculate this interference pattern in the plane wave approximation.

The net \( \mathbf{E} \) field resulting from interference of six plane waves of frequency \( \omega \) with wave vectors
\[
\mathbf{k}_{\pm x} = \pm k\hat{x}, \quad \mathbf{k}_{\pm y} = \pm k\hat{y}, \quad \mathbf{k}_{\pm z} = \pm k\hat{z},
\] (D.1)

amplitudes \( E_j \) and arbitrary offset phases \( \varphi_j \) is given by the sum
\[
\mathbf{E}(\mathbf{r}, t) = \sum_{j=\{x\pm,y\pm,z\pm\}} \hat{e}_j E_j e^{i(k_j \cdot \mathbf{r} + i\varphi_j - i\omega t)} + c.c.
\] (D.2)

Summation index refers to the relevant axes of the reference frame and directions of the wave propagation, indicated by the \( \pm \) sign. Polarization unit vectors of the corresponding waves are denoted as \( \hat{e}_j \). Polarization of the incident MOT beams is characterized by the set of unit vectors
\[
\hat{e}_{x\pm} = \frac{1}{\sqrt{2}} (\hat{y} \pm i\hat{z}), \quad \hat{e}_{y\pm} = \frac{1}{\sqrt{2}} (\hat{z} \pm i\hat{x}), \quad \hat{e}_{z\pm} = \frac{1}{\sqrt{2}} (\hat{x} \mp i\hat{y}).
\] (D.3)

Assuming that all incident waves have the same amplitudes \( E_j = E_0 \), we obtain
\[
\mathbf{E}(\mathbf{r}, t) = \frac{E_0}{\sqrt{2}} \begin{pmatrix}
\cos(kz + \varphi_{x+} - \omega t) + \cos(kz - \varphi_{z-} + \omega t) \\
-\sin(ky + \varphi_{y+} - \omega t) - \sin(ky - \varphi_{y-} + \omega t) \\
\cos(kx + \varphi_{x+} - \omega t) + \cos(kx - \varphi_{x-} + \omega t) \\
+\sin(kz + \varphi_{z+} - \omega t) + \sin(kz - \varphi_{z-} + \omega t) \\
\cos(ky + \varphi_{y+} - \omega t) + \cos(ky - \varphi_{y-} + \omega t) \\
-\sin(kx + \varphi_{x+} - \omega t) - \sin(kx - \varphi_{x-} + \omega t)
\end{pmatrix}.
\] (D.4)

The way in which \( \mathbf{E}(\mathbf{r}, t) \) depends on phases \( \varphi_j \) has significant consequences. For example, when we set \( \varphi_j = 0 \) for each \( j \), we obtain a simple expression for \( \mathbf{E}(\mathbf{r}, t) \), namely
\[
\mathbf{E}(\mathbf{r}, t) = \frac{2E_0}{\sqrt{2}} \begin{pmatrix}
\cos(kz) - \sin(ky) \\
\cos(kx) + \sin(kz)
\end{pmatrix} \cos(\omega t).
\] (D.5)
From equation (D.5) it follows that in each point the net field polarization is linear with spatially modulated direction. Hence, the case when \( \varphi_j = 0 \) can be treated as a generalization of the 1D \( \sigma^+ - \sigma^- \) beams configuration. The main difference between the 1D and 3D situations is modulation of the light intensity in the 3D case, depicted in Figure D.1. The intensity is calculated according to equation

\[
I = C\langle \mathbf{E}(\mathbf{r}, t) \cdot \mathbf{E}(\mathbf{r}, t)^* \rangle = C \int_0^{2\pi} E(\mathbf{r}, t) \cdot E(\mathbf{r}, t)^* \, dt. \tag{D.6}
\]

In this Appendix, we set \( C = 1 \), hence \( I \) is given in arbitrary units.

To see how the topology of the interference pattern changes when the beam phases are altered, we set \( \varphi^+ = \frac{\pi}{2} \) and then \( \varphi^- = \pi \). The results are depicted in Figures D.2 and D.3, respectively. Now, both intensity and polarization are modulated. Polarization is elliptical in local intensity maxima and linear in minima. Also, unlike in the previously discussed case, the intensity modulation does not reach 100% contrast.

The results indicate that fluctuations of the trapping beam phases (Appendix C) result in a time varying, random realization of the net field. Hence, our experimental results can be treated as the spectral response of the system averaged over these realizations. Because of randomness of the net field, no stable atomic localization and optical lattices are expected.
Figure D.1: Light intensity modulation in the plane $z = \frac{3}{8}$ for all $\varphi_j$ equal to 0. Light polarization is linear everywhere but its direction is space-dependent. In the local intensity maxima, i.e. in points $A = \left(\frac{1}{8}, \frac{3}{8}, \frac{3}{8}\right) \lambda$ and $B = \left(\frac{7}{8}, \frac{9}{8}, \frac{3}{8}\right) \lambda$, the net intensity is $I = 6|E_0|^2$. In the intensity minima, i.e. in points $C = \left(\frac{5}{8}, \frac{5}{8}, \frac{3}{8}\right) \lambda$ and $D = \left(\frac{3}{8}, \frac{7}{8}, \frac{3}{8}\right) \lambda$, the intensity drops to zero.

Figure D.2: Light intensity modulation (a) and polarization (b) in the plane $z = 0$ for $\varphi_{z+} = \frac{\pi}{2}$, $\varphi_{j\neq z+} = 0$. Light polarization is elliptical at the intensity maxima and linear at minima, as shown in plots (b). At local intensity maxima, for $A = \left(\frac{1}{8}, \frac{7}{12}, 0\right) \lambda$ and $B = \left(\frac{5}{8}, \frac{11}{12}, 0\right) \lambda$, $I = 5.5|E_0|^2$. At minima for $C = \left(\frac{1}{4}, \frac{13}{12}, 0\right) \lambda$ and $D = \left(\frac{1}{6}, \frac{5}{12}, 0\right) \lambda$, $I = 0.5|E_0|^2$. Axes of plots (b) are in $E_0$ units.
Figure D.3: Light intensity modulation (a) and polarization (b) in the plane $z = 0$ for $\varphi_{z+} = \pi$, $\varphi_{\delta \neq z+} = 0$. Light polarization is elliptical at the intensity maxima and linear at minima, as shown in plots (b). At local intensity maxima, for $A = (\frac{1}{4}, \frac{1}{4}, 0)\lambda$ and $B = (\frac{3}{4}, 1, 0)\lambda$, $I = 5|E_0|^2$. At minima for $C = (\frac{1}{4}, 1, 0)\lambda$ and $D = (\frac{3}{4}, 1, 0)\lambda$, $I = |E_0|^2$. Axes of plots (b) are in $E_0$ units.
Bibliography


