THERMAL PLASMA DIAGNOSTICS USING DEGENERATE FOUR-WAVE MIXING

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The non-intrusive degenerate four wave mixing (DFWM) method was shown to be useful in diagnostics of the thermal equilibrium arc plasma. The thermal atmospheric-pressure argon arc plasma was studied and the laser wavelength was in resonance with the 4s[3/2]^2-4p'[1/2] ArI transition, corresponding to the 696.5nm emission line. The Abrams-Lind theory was verified and proved to be valid under conditions of our plasma. In the high-laser intensity limit, the DFWM signals were shown to be exclusively dependent on the population difference between the relevant argon states. Well resolved axial profiles of the plasma temperature and the electron density were then calculated.

1. Degenerate Four Wave Mixing

Degenerate four-wave mixing (DFWM) is a process where three electromagnetic (EM) waves of the same frequency ω interact with a nonlinear medium to form the fourth, phase-conjugated, EM signal wave. Physically DFWM can be viewed as Bragg scattering of EM wave from low-frequency plasma density modulation formed by the coupling of two EM waves through plasma nonlinearity. Scattering of the third EM wave from this density grating yields the signal wave. The signal wave is generated in the direction strictly determined by the phase-matching conditions that are simply momentum and energy conservation:

\[ k_f + k_b = k_p + k_s, \omega_f + \omega_b = \omega_p + \omega_s \]

where \(k_i\) and \(\omega_i\) stand for wave vectors and frequencies of EM waves, respectively.

In our work we applied DFWM to study the thermal equilibrium arc plasma and a schematic diagram is depicted in Fig. 1. Two pump beams with electric vectors \(E_f\) and \(E_b\) are co-axial and counter-propagating. The third beam \(E_p\) crosses the pump beams axis at an angle \(\theta\). All three laser beams of the same frequency \(\omega\) couple through interaction with the nonlinear medium to generate the fourth beam \(E_s\) that propagates exactly opposite to and collinear with \(E_p\). This geometry satisfies the phase-matching condition for all angles \(\theta\). The interference of two laser beams results in a spatial light-intensity modulation pattern with fringe spacing

\[ \Lambda = \lambda / 2 \sin (\theta / 2) \]

where \(\lambda\) is the laser wavelength. The spatial oscillation of the light intensity results in a similar variation of the concentration of the upper and lower states of the optical transition. This, in turn, makes a refraction and absorption coefficients to vary and form the Bragg
grating. The diffraction gratings produced by the interference of \( E_p \) with \( E_f \) and \( E_p \) with \( E_b \) results in scattering of \( E_b \) and \( E_f \) respectively, and generation of the signal beam \( E_s \).

![Diagram](image)

**Fig. 1.** (A) Diagram of the degenerate four wave mixing (DFWM) geometry; (B) Schematic drawing of the grating formed by the interference of the forward and the probe laser beams

The DFWM process is usually described by a nonlinear polarizability of the medium and Maxwell's Equations which lead to the wave equation,

\[
\nabla^2 E - \frac{1}{c^2} \left( \frac{\partial^2 E}{\partial t^2} \right) = \frac{4\pi}{c^2} \frac{\partial^2 P}{\partial t^2}; P = P^{(1)} + P^{NL}
\]

The nonlinear polarizability can be written in the form

\[
P_i = \chi^{(1)}_{ij} E_j + \chi^{(2)}_{ijkl} E_k E_l + \chi^{(3)}_{ijkl} E_k E_l E_j E_i + ...
\]

where \( \chi(E) \) stand for nonlinear susceptibilities. If there are several EM waves involved, then

\[
E = E_a + E_b + E_c + ...
\]

The solution of the above equations yields the signal EM wave \( E_s \) amplitude

\[
E_s = \chi^{(1)}(E) \left[ (E_f \cdot E_p^*) E_b + (E_b \cdot E_p^*) E_f \right],
\]

As seen in (1.6), \( E_s \) is conjugated to \( E_p \) which illustrates phase - conjugation of this kind of DFWM. In general, the signal depends also on a relative polarization of the initial waves.

The model for the DFWM in absorbing, homogeneously broadened two-level media has been worked out by R.L. ABRAMS et al. [1, 2]. The Abrams-Lind (A-L) model assumes undepleted pump beams \( (I_f, I_b) \) and allows small absorption for the probe \( (I_p) \) and signal \( (I_s) \) beams. Furthermore, assumptions of monochromatic laser fields with equal pump beam intensities \((I_p = I_b = 1)\) and of a low \( I_p \) intensity \((I_p << I_{sat})\) in the low and high-laser intensity limits yield:

\[
I_s \propto \Delta N_0^2 L^2 \mu^8 T_1^2 T_2^4 \frac{P^3}{(1 + 8^2)^3}; \quad (I << I_{sat}),
\]
\[ I_s \propto \Delta N_0^2 L^2 \mu^2 \frac{T_2}{T_1}; \quad (I \gg I_{sat}) \]

where: \( \Delta N_0 \) is the population difference between levels of the atomic transition in the absence of laser fields, \( \mu \) is the transition dipole moment, \( \delta \) is the laser detuning from the atomic transition frequency \( \omega_0 \), normalized to the atomic transition linewidth, \( I_{sat} \) is the saturation intensity:

\[ I_{sat} = \left( \frac{e_0 c \hbar^2}{2 T_1 T_2 \mu^2} \right) (1 + \delta^2) = I_{sat}^0 (1 + \delta^2) \]

where \( T_1 \) and \( T_2 \) are the population and coherence decay times, respectively and \( e_0 \) stands for the vacuum permittivity.

At the low-intensity limit, the signal line shape \( I_s \) is proportional to the Lorentzian profile of an atomic spectral line in the third power, and the signal scales like the third power of the laser intensity. For the strong-intensity (saturation regime), the DFWM signal is insensitive to the laser intensity.

The latter case can be viewed as very useful for direct monitoring of a population difference in the medium. Furthermore, when the lower level is a ground level, the DFWM signal is practically proportional to a total concentration of probed species.

In comparison to optical emission spectroscopy the DFWM method gives coherent, highly collimated signals of high spatial resolution.

2. Argon Arc Plasma Diagnostics Using DFWM

The A-L theory of the DFWM in its high-laser intensity limit gives the simple formula equation (1.8), especially useful in the case of the LTE plasma. Signal sensitivity on the relative dephasing rates \( (T_2/T_1) \) is greatly reduced as a result of a domination of electron collision processes in the plasma where both \( T_1 \) and \( T_2 \) are directly proportional to the electron density, so their ratio has a constant value throughout the studied plasma volume. Thus spatial variation of DFWM signals can be predominantly attributed to the variation in the population difference \( \Delta N_0 \), enhanced by a quadratic dependence shown in equations (1.7) and (1.8). This population difference can be calculated for an electric argon arc plasma column J.B. SHUMACKER et al. [11], H. NUBBEMEYER [5], W. NEUMANN [4], H.R. GRIEM [3], S. PELLERIN et al. [7] using LTE plasma model and this way spatial distribution of plasma parameters (electron temperature and density) is determined.

LTE plasma model assumes a Maxwellian velocity distribution with the same temperature \( T_e \) for all plasma components. The populations of atomic and ionic excited states are described by the Boltzmann distribution while the Saha equation describes the density of different plasma elements in two consecutive ionization stages (e.g. atoms, ions and free electrons). In the frame of the LTE model, the population difference \( \Delta N_0 \) between any two atomic states in the plasma is calculated at given plasma temperature \( T_e \), and pressure \( p \), and for singly ionized plasma is given by

\[ \Delta N_0 = \frac{N_e^2}{S_0} \left[ \exp(-E_1 / k_B T_e) - \exp(-E_2 / k_B T_e) \right] \]
where: \( N_e \) is a free electron density, \( E_1, E_2 \) – energies of the lower and upper states, \( k_B \) – the Boltzmann constant and \( S_0 = S_0(T_e) \) is the Saha function for this atomic component W. NEUMANN [4], H.R. GRIEM [3].

We define the norm population difference \( \Delta N_0^N \) as a maximum value of \( \Delta N_0 \), attainable at the specific temperature \( T_e^N \) (called the norm temperature) for the LTE atmospheric pressure plasma. These terms are introduced in a similar way as the norm intensity and temperature for spectral lines W. NEUMANN [4]. The population difference \( \Delta N_0 \) between two specific argon states (4s[3/2] and 4p’[1/2], 696.5nm ArI line), calculated for the LTE atmospheric pressure plasma, versus the plasma temperature is plotted in Fig. 2 with the solid line. The norm population difference and the appropriate norm temperature have values of \( \Delta N_0^N = 1.506 \times 10^{19} \text{m}^{-3} \) and \( T_e^N = 14900 \text{K} \), respectively and are marked in the figure. For the sake of completeness, the temperature dependence of free electron density (under the LTE plasma model) is also plotted in Fig. 2 with the dashed curve. Once the norm temperature has been reached, and \( \Delta N_0^N \) measured at some place in the investigated plasma volume, \( \Delta N_0 \) can be derived at any plasma place, \( x \), from the relation:

\[
\Delta N_0^N (x) = \Delta N_0^N \sqrt{I_s (x) / I_s (N)} ,
\]

where \( I_s (x) \) and \( I_s (N) \) are the DFWM signals measured at the position \( x \) and at position of the maximum (norm value) \( \Delta N_0^N \), respectively. The plasma temperature and electron density can be calculated using the LTE plasma equation set W. NEUMANN [4].

![Fig. 2. The population difference \( \Delta N_0 \) (solid curve) between the argon states 4s[3/2] and 4p’[1/2] and the electron density \( N_e \) (dashed curve) versus plasma temperature \( T_e \) calculated under LTE atmospheric pressure plasma conditions.](image)
3. Experimental Set-up

The experimental arrangement consists of the studied plasma source, the laser and the detection system. The plasma source has been described in detail in our previous publications B. POKRZYWKAV. et al. [9], or PELLERIN et al. [7], for example - see too paper named "Study of the cathodic zone of an electric arc" (by B. Pokrzywka, S. Pellerin, K. Musiol, K. Dzierżega, E. Pawelec and J. Chapelle) in this review. Briefly, the arc discharge is generated from a tungsten conical cathode tip (α=40° conical angle) of 4mm diameter and surrounded by a water cooled nozzle. In its upper part, the arc consists of three copper discs with a 5mm channel in the center. The third disc serves as anode. The arc is operated in pure argon and its flow of 2.0l/min is maintained. The arc is powered by current ranging from 70 to 120A.

A tunable dye laser (λ near 696.54nm) is pumped by a second harmonic of a Nd:YAG laser with a 5Hz repetition rate. The dye laser provides 10ns pulses of about 600μJ energy with a spectral bandwidth less than 9GHz. The output beam of the dye laser is split into a forward-pump \( I_f \) and a probe \( I_p \) beams with an intensity ratio of 8:1. The forward beam after passing plasma region is reflected from a dielectric concave mirror back into the plasma interaction region and acts as a third, backward-pump beam \( I_b \). The 500mm focal length lens and the 200mm focal length mirror are used to focus the laser beams at the plasma symmetry axis with a waist diameter smaller than 0.1mm.

In a phase-conjugate geometry of Fig. 1, two counter propagating pump beams cross with the probe beam at an angle of ~14° which determines a spatial resolution of the method to be: 0.1mm longitudinally and 0.35mm transversally with respect to the laser beam. In our experiment we employ a crossed polarization configuration in which a quarter-wave plate (WP) is used in order to rotate the polarization of the backward-pump beam 90° with respect to that of the horizontally polarized forward-pump and probe beams.

The generated DFWM signal \( I_s \) with a same polarization as the backward-pump beam propagates backward along a probe beam path and is directed towards the detection system. The signal detection is performed by a fast photomultiplier (PMT) installed behind the exit slit of a monochromator. The PMT signal is then directly connected to a digital oscilloscope (300MHz bandwidth), externally triggered by the YAG laser pump light pulses.

The radial and horizontal profiles of the DFWM signal are obtained by translating of the plasma arc with a step-motor drive. The whole acquisition system is operated and controlled by a computer.

4. Results

Figure 3 presents the DFWM signal versus the laser intensity obtained for the argon plasma discharge operated at 90A and measured at the plasma symmetry axis (R=0mm), 1mm above the cathode tip. The solid curve is a theoretical fit to experimental data according to equation (1.7) and (1.8). This figure shows that we reached the saturation limit and the A-L theory correctly describes our signals.
The population difference $\Delta N_0$, according to equation (1.8), is derived as a square root of the DFWM signal. Its axial variations, for the plasma operated at "low" (70A) and "high" (120A) current, are shown in Fig. 4. In both cases, $\Delta N_0$ exhibit a maximum at a distance of 2,35mm and 3,6mm above the cathode for low and high current case, respectively.

From our previous optical emission spectroscopy (OES) measurements S. Pellerin et al. [7], we know that for this plasma source the temperature on the arc axis grows toward the cathode and the LTE state exists in the near axis plasma zone.

![Graph showing comparative DFWM signal dependence on laser intensity](image)

**Fig. 3.** Comparison of the experimental (■) and theoretical (→) DFWM signal dependence on the laser intensity. The theoretical curve are calculated according to the A-L theory (Eq.8) for the resonance excitation ($\delta=0$).

Therefore, from Fig. 4, we conclude that the axial maximum of $\Delta N_0$ corresponds to the norm value defined in part 3, with the norm temperature $T_e^N = 14900$K and free electron density $N_e=1.635\times10^{23}$m$^{-3}$.

The axial temperature and electron density distributions are determined as described above and are shown in Fig. 5. The solid and open symbols stand for the plasma temperature and electron density while circles and squares for low and high arc current, respectively. This figure displays an axial temperature increase in the cathode direction. For the higher current the temperature remains about 500 K higher than for the lower one, all along the plasma axis.
Fig. 4. Axial distribution of the population difference $\Delta N_0$, obtained from the DFWM signals measured in the high laser intensity limit [Plasma arc current: (●) - $I_{arc}=70\text{A}$; (■) - $I_{arc}=120\text{A}$] ($H=0\text{mm}$ corresponds to the cathode tip).

Fig. 5. Axial distributions of the plasma temperature (solid symbols) and electron density (open symbols), determined from the DFWM signals as described in §3. [Plasma arc current: (●, ○) - 70A; (■, □) - 120A] ($H=0\text{mm}$ corresponds to the cathode tip).
Unlike the temperature, close to the electrode (up to 1.25mm), the electron density exhibits its maximum value under the LTE atmospheric plasma conditions (see Fig. 2) and shows no variation with a further electrode approach. On the other hand, above 1.5mm, $N_e$ rapidly decreases with a distance from the electrode.

5. Conclusion

Finally, the non-intrusive degenerate four wave mixing (DFWM) method was shown to be useful in diagnostics of the thermal equilibrium arc plasma. It was used to study the LTE atmospheric pressure argon plasma, and the laser wavelength was in resonance with the 4s[3/2]$^4$P$^o$-4p'[1/2] ArI transition, corresponding to the 696.5nm emission line. The occurrence of a deviation from the LTE plasma model near the cathode tip B. POKRYWKA et al. [9, 10], PELLERIN et al. [6-8], despite high temperature and electron density.

The main advantages of the DFWM are due to its relatively simple and fast data processing. Particularly, the measured signal is directly proportional to the atomic transition population difference $\Delta N_0$. Furthermore, useful information (plasma temperature, local component densities...) may be obtained for asymmetrical plasmas, contrary to the OES measures, which require the cylindrical symmetry hypothesis to achieve an Abel inversion.

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References