Measurements of Stark widths and shifts of Ne I lines using degenerate four-wave mixing and Thomson scattering methods

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Abstract

We report on measurements of Stark widths and shifts of four prominent Ne I lines of the 3s,3s′-3p transition arrays. The measurements were performed in an atmospheric-pressure arc discharge operated in argon–neon gas mixture. Sub-Doppler degenerate four-wave mixing technique was used to measure the line profiles, while Thomson scattering yielded the plasma parameters: electron density, \(n_e=(0.53–1.33) \times 10^{23} \text{ m}^{-3}\), and electron temperature, \(T_e=10,200–20,900 \text{ K}\). The measured profiles are symmetric within the uncertainty limits. The experimental Stark widths and shifts are compared with results of other experiments and theoretical calculations. © 2006 Elsevier B.V. All rights reserved.

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1. Introduction

The development of space born astronomical techniques and devices such as Goddard high-resolution spectrograph on the Hubble space telescope has increased the demand for atomic data of many elements. Neon is the fifth abundant element in the universe after hydrogen, helium, oxygen and carbon, and it constitutes the interiors of many stars and white dwarfs. The spectral lines of neutral neon are particularly abundant in low-temperature (10000–30000 K) astrophysical and laboratory plasmas. In such kind of plasmas (with electron densities \(10^{21}–10^{24} \text{ m}^{-3}\)), the Stark broadening is the principal pressure broadening mechanism of the spectral lines and is of great interest for plasma diagnostics purpose.

In a number of experimental studies [1–10], the Stark widths and shifts of Ne I non-resonance lines were reported and they were compared with semiclassical calculations [11]. For the Stark widths, the agreement between theory and experiment is on average within 20% [12,13]. Much larger discrepancies were reported in [3] and they were ascribed to the uncorrected effect of self-absorption [5,12]. For the Stark shifts, the discrepancies between theory and experiment are larger than for the Stark widths and in some cases reach a factor of 2 [4]. Recently, the measurements of the ion-broadening parameters \(A\) of several Ne I lines were reported [7–9]. The values of \(A\) were found to be 3 to 15 times larger than predicted in [11] and they strongly disagree with the results of the former experiments where the symmetric (within the uncertainty limits) line profiles were measured.

In all of these experiments but [5], the low-pressure pulsed discharge was used as the excitation source. The electron density was deduced from the measurements of the Stark widths of hydrogen or helium lines or from laser interferometric data, while the total intensities of the spectral lines were measured to determine the electron temperature. The line profiles were studied using optical emission spectroscopy (OES).

The objective of this paper is to resolve the above-mentioned discrepancies using the alternative laser-based experimental techniques: degenerate four-wave mixing (DFWM) and Thomson scattering (TS) and the atmospheric-pressure dc arc discharge as...
the excitation source. DFWM was used to measure the Stark profiles, while TS served for plasma diagnostics.

The DFWM method is characterized by high spatial resolution, which is determined by the overlap volume of the involved laser beams. Moreover, in the backward (phase-conjugate) geometrical configuration, DFWM also offers superior spectral resolution as compared to OES owing to its sub-Doppler character. The latter is especially important in the case of light elements like neon. The DFWM method has been recently applied to study the Stark profile of the 696.5 nm Ar I line in an atmospheric-pressure argon arc plasma [14]. Principal advantages of Thomson scattering are also high spatial resolution and ease with which the measured data can be interpreted. Indeed, in contrast to many other techniques, derivation of the standard plasma parameters, the electron temperature and density, does not require any assumptions about the plasma symmetry, equilibrium state or its chemical composition.

The local character of DFWM and TS makes these methods very suitable to study Stark broadenings in plasmas for which the assumptions of homogeneity and symmetry are always questionable and where the Doppler broadening and the cold boundary layers can significantly distort the measured profiles.

2. Experimental arrangement and measurements

The cross-sectional view of the plasma generator is depicted in Fig. 1. The arc discharge was generated from a conical cathode tip (cone angle of 60°) made of a 2-mm diameter, thoriated (2%) tungsten rod surrounded by a water-cooled nozzle. The upper part of the arc consisted of two copper discs with a channel of 5-mm diameter. The first disc served to improve the arc stability, while the second disc was used as the anode. The distance between the cathode tip and the first disc was about 9 mm. The arc was operated at atmospheric pressure in a gas mixture of argon and neon with variable partial pressures of Ne/Ar from 2:3 to 2:1. The total gas flow was kept at 4 l/min in a gas mixture of argon and neon with variable partial pressures was about 9 mm. The arc was operated at atmospheric pressure anode. The distance between the cathode tip and the first disc improve the arc stability, while the second disc was used as the nozzle. The upper part of the arc consisted of two copper discs

![Fig. 2. Geometry of the Thomson scattering: $\mathbf{k}_T$ is the wave vectors of the incident laser beam and the scattered light, respectively, while $\mathbf{k} = \mathbf{k}_T - \mathbf{k}_s$ is the scattering wavevector. $\theta$ is the scattering angle. The polarization of the incident laser beam is vertical, i.e. laser field $E_T$ is perpendicular to the scattering plane ($\mathbf{k}_e$, $\mathbf{k}_T$).](image)

axis was probed with the laser beams. Depending on discharge conditions and the studied plasma region above the cathode tip, plasma was characterized by $n_e = (0.53 - 1.33) \times 10^{23} \text{ m}^{-3}$ and $T_e = 10,200 - 20,900 \text{ K}$.

2.1. Plasma diagnostics

Plasma diagnostics was performed by measuring the Thomson scattering of laser radiation of a vertically polarized second harmonic ($\lambda = 532 \text{ nm}$) of a Nd:YAG laser which delivered pulses of energy of 55 mJ and of about 6 ns duration. The beam was focused to the spot with the diameter of about 300 μm on the plasma axis and a few millimeters above the cathode tip. The scattered light was collected at the angle $\theta = 80.9^\circ$ (see the configuration in Fig. 2). The investigated plasma volume was imaged onto the entrance slit of a spectrograph (1.5 nm/mm reciprocal dispersion) with magnification equal to 1. The width of the slit was set to 20 μm. The Thomson scattered spectra were collected over the wavelength range of 521.9–542.1 nm. In order to improve the signal-to-noise ratio, only the vertically polarized signal was selected by inserting a polarizer in the path of the scattered light. Single-shot scattering spectra were measured using a gated intensified two-dimensional charge-coupled device (ICCD) camera and averaged over 1000 laser shots. The TS spectra were obtained by subtracting the spectra collected with and without the laser beam. The spectral and spatial resolutions were found to be 0.03 nm and 24 μm, respectively. The camera was synchronized to the laser pulse with variable delay and the time resolution was limited to 5.0 ns by the minimal gate width of the camera.

Under our experimental conditions of low-temperature ($T_e \approx 15,000 \text{ K}$) and high-density ($n_e > 10^{22} \text{ m}^{-3}$) plasma, the TS spectrum has a partially collective character [15], which enables to independently determine both $n_e$ and $T_e$ without absolute calibration of the scattered spectra [15–18].

However, due to very low scattering cross-section for the TS process and strong plasma radiation background, high laser powers are required to obtain a detectable TS signal. Unfortunately, high laser power can substantially perturb the plasma state by absorption of laser radiation predominantly in the inverse bremsstrahlung (IB) process. These effects have been recently studied in [15,19,20]. Under conditions of our experiment, the electron density was not affected by the laser pulse, which was
verified by studying the spatial variations of \( n_e \) within the laser beam as it was suggested in [15]. Unlike \( n_e, T_e \) was changing with position within the laser beam so that its initial, “undisturbed”,
value could not be directly derived. Moreover, because of the insufficient temporal resolution of the experiment, it was impossible to correct for the heating effect by studying the temporal evolution of \( T_e \) during the laser pulse and then by extrapolating the results to the origin of the pulse [15].

Instead, we measured the TS spectra during the early stage (about first 3 ns) of the laser pulse. Next, based on the derived electron density and temperature, the initial value of \( T_e \) was calculated using the formula for the upper limit for the laser-induced increase of \( T_e \) [16,21,15]

\[
\Delta T_e = \frac{2}{3} \frac{k_B n_e}{\hbar^2} E_h T_e, \tag{1}
\]

where \( E_h \) is the laser energy and \( r_0 \) is the laser beam radius in the plasma region. The absorption coefficient \( \kappa_{IB} \) for IB is given by

\[
\kappa_{IB} = \left( \frac{\pi Z^2}{2 \lambda} \right)^{1/2} \frac{n_e Z^2}{m_e c^2} \left( \frac{\pi \omega_0}{4 \pi \omega_1} \right)^{3/2} \frac{\hbar^3}{m_e} \left( \frac{\omega_0}{\omega_1} \right)^{1/2} n_e n_z (1-\exp(-\hbar \omega_1 / \hbar \omega_1)) \frac{\omega_{RF}}{\omega_1}, \tag{2}
\]

where \( Z \) is the ion charge, \( n_{e,z} \) is the density of ions in the \( Z \)-th ionization stage and \( \omega_{RF} \) denotes the Gaunt factor for free–free transitions. In our calculations, we assumed the square 6 ns long laser pulse and singly ionized plasma. This procedure gave the initial, “undisturbed” electron temperatures several thousands Kelvins lower than the ones directly measured.

The uncertainties of the electron density do not exceed 3% and are of statistical origin. The uncertainties of \( T_e \) are 5–11% and reflect mainly the errors of the corrections for the heating effect.

2.2. Stark profile measurements

In parallel to the TS measurements, the line profile of the investigated transition was registered using the DFWM method.

DFWM involves three laser beams of identical optical frequencies \( \nu_1 \) which interact through the nonlinearity of a medium to generate a fourth, signal beam, at the same frequency. We applied DFWM in the backward phase-conjugate (PC) geometry as it is shown in Fig. 3. The probe beam (\( I_p \)) crosses the two counter-propagating forward (\( I_0 \)) and backward (\( I_b \)) pump laser beams at an angle \( \theta_{0P} \). The generated signal beam (\( I_s \)) has the same optical frequency \( \nu_1 \) and is phase-conjugated to the probe beam. In other words, it propagates backward with respect to the probe beam. In the PC configuration, the influence of the Doppler effect on the DFWM line profile is strongly reduced because the atoms resonantly interacting with all laser beams contribute most significantly to the signal.

Detailed discussion of the theory of DFWM is presented in several monographies [23,22,24]. In the case of stationary two-level systems and in the limit of non-saturating laser beams, the spectral profile of the signal beam is given by

\[
I_s(\delta) = C \left( \frac{1}{1 + \delta^2} \right)^3 \frac{I_0 I_b I_s}{I_{sat}}, \tag{3}
\]

where \( \delta = (\nu_1 - \nu_0) / \omega \) is laser detuning from the resonance frequency \( \nu_0 \) of the system expressed in units of the homogeneous line width (\( \omega \)). \( I_{sat} \) is the saturation intensity, \( C \) includes all physical constants and decay rates characterizing the system at given experimental conditions.

The Stark-broadened line shapes for PC-DFWM laser spectroscopy have been recently studied in [25]. The theoretical model of the PC-DFWM spectral profile takes into account the contribution of the Stark effect together with the ion dynamics and the Doppler effect as well as the geometrical configuration of the laser beams. Calculations were performed in the limit of low laser intensities, for high-density \( \left( n_e > 10^{21} \text{ m}^{-3} \right) \) and low-temperature \( \left( T_e \approx 10,000 \text{ K} \right) \) plasma conditions. The resultant PC-DFWM profiles are shown to be significantly less broadened than emission profiles though they both are similarly shifted. In \( w_0 < w \) limit, the measured width \( w_{dfwm} \) approaches \( w/2 \) (see [25]). Furthermore, the asymmetry due the static ion broadening is somewhat less pronounced in the PC-DFWM profile than in the emission profile. Finally, calculations show a great effect of the geometric configuration of the laser beams on the PC-DFWM line profile as long as the Doppler broadening is significant. Fig. 4 shows the relative linewidth of the PC-DFWM signal \( w_{dfwm}/w \) calculated within the model presented in [25] versus the Doppler broadening \( w_D/w \) and for different angles \( \theta_{0P} \) between the forward pump and the probe laser beams. As one can observe, even for \( w_{DF} \gg w \), the influence of

![Fig. 3. Geometric configuration of the backward phase-conjugate DFWM.](image-url)

![Fig. 4. Dependence of the linewidth \( w_{dfwm} \) measured with DFWM method on the Doppler broadening \( w_0 \) measured in units of the homogeneous broadening \( w \). Calculations performed for different angles \( \theta_{0P} \) between the pump and probe laser beams, (●) 2°, (○) 5°, (▼) 20°, (◇) 45°.](image-url)
the Doppler broadening on the PC-DFWM profile can be greatly suppressed provided the collinear configuration of the laser beams is used. With these calculations, it is possible to correct the Stark profiles measured with the DFWM method for the Doppler broadening.

In our experiment, a tunable dye laser was pumped by the second harmonic of another Nd:YAG laser with 10 Hz repetition rate. The dye laser provided 8 ns laser pulses with a spectral bandwidth of less than 0.05 cm$^{-1}$ and energies of about 2 mJ. The output beam of the dye laser was split into three beams with intensities $I_f/I_b/I_p = 4:4:1$ and with the backward pump beam polarized perpendicularly to the other two beams. The 400-mm focal length lenses were used to focus the laser beams at the plasma symmetry axis with a waist diameter of about 100 $\mu$m. Two counter-propagating pump beams intersecting with the probe beam at the angle $\theta_{fp} \approx 20^\circ$ determined the spatial resolution of the measurements to be at least 100 $\mu$m longitudinally and 200 $\mu$m transversally. The generated PC-DFWM signal propagated backwards along the probe beam path. It was then separated from the probe beam by a polarizing beam splitter cube. The prism monochromator with the spectral bandwidth set to 1 nm discriminated the signal from the plasma radiation background. The signal was detected with a fast gated photomultiplier and then amplified with the pre-amplifier and recorded on a digital oscilloscope (300 MHz bandwidth). Signals were averaged over 10 to 50 laser shots and then integrated over a time period of 15 ns. The PC-DFWM spectrum was simultaneously recorded with the reference optogalvanic spectrum of a neon hollow cathode (HC) lamp and the intensity of the dye laser was monitored with a fast photodiode. The details of the experimental setup can be found in [14].

The laser beams for TS and DFWM measurements were overlapped to probe the same plasma volume. However, the dye laser and the Nd:YAG laser pulses used for DFWM and TS measurements were separated in time by several hundreds nanoseconds in order to avoid any possible impact of one measurement on another.

3. Results and discussion

The Stark widths and shifts of four prominent Ne I lines belonging to the $3s,3s' -3p$ transition arrays were measured. For each line, two separate experiments were carried out. In the first experiment, the Stark profile (width and shift) was studied at particular plasma conditions ($n_e$ and $T_e$). The line broadening by laser radiation was eliminated by decreasing the laser power until no effect on the line profile was observed. The absorption of the signal beam in the plasma was found negligible which results from short optical path (about 5 mm long) travelled across the plasma column by the signal beam. The line shifts were determined with respect to the optogalvanic signal registered from a low-pressure neon HC lamp. The symmetric (within the uncertainty limits) line profiles were measured for all investigated transitions and Fig. 5 shows an example of such profile for the 640.22 nm Ne I line. These symmetric profiles contradict the findings reported in [7–9] and this discrepancy might be related to some uncontrolled inhomogeneities of their plasma column (see [10]).

The line width $w_{dfwm}$ and the Stark shift $d$ were determined by fitting the experimental data with Eq. (3). The Stark width $w$
was then derived correcting the measured linewidth $w_{\text{dfwm}}$ for the Doppler broadening with the use of the results of numerical calculations [25] presented in Fig. 4. Under our experimental conditions, the Doppler broadening $w_D = 0.3 \text{ cm}^{-1}$ and $w_D/w_{\text{dfwm}} \approx 0.4$, the measured linewidths were found to be enlarged by about 15% with respect to the Stark widths. Although the two-temperature ($T_g < T_e$) model for our plasma should be considered, for these calculations, we assumed $T_g = T_e$. Indeed, lowering the gas temperature, $T_g$ by as much as 5000 K resulted in the increase of the final Stark width only by less than 1.5%.

The second experiment studied the dependence of the Stark shifts on plasma parameters ($n_e$ and $T_e$). These measurements were performed at higher laser intensities than in the first experiment in order to increase the S/N ratios especially for plasma conditions characterized by lower electron densities and thus weaker DFWM signals. The plasma conditions were altered by changing either Ne/Ar concentration in the gas mixture, the arc current or the investigated plasma region above the cathode. For the studied temperature range and within the experimental uncertainties, no correlation between the normalized ($n_e = 10^{23} \text{ m}^{-3}$) Stark shifts and $T_e$ could be established; the results for the 640.22 nm Ne I line are plotted in Fig. 6. The normalized Stark shifts $d_s$ for $n_e = 10^{23} \text{ m}^{-3}$ were obtained assuming the linear dependence between the measured Stark shift and electron density (see Fig. 7).

In Tables 1 and 2, the normalized Stark widths $w_{\text{N}}$ and shifts $d_s$ are listed together with the results of other experiments. In these tables, we also compare measured Stark widths and shifts

### Table 1
The Ne I Stark widths (FWHM) $w_{\text{N}}$ in cm$^{-1}$ normalized to $n_e = 10^{23} \text{ m}^{-3}$

<table>
<thead>
<tr>
<th>Transition, $\lambda$ (nm)</th>
<th>$n_e$ ($10^{23} \text{ m}^{-3}$)</th>
<th>$T_e$ (K)</th>
<th>$w_{\text{N}}$ (cm$^{-1}$)</th>
<th>$w_{\text{N}}/w_{\text{in}}$</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>3s$^{3}[3/2]_0 – 3p[3/2]_1$</td>
<td>0.67–0.88 33,000–36,500</td>
<td>3.3</td>
<td>0.76–0.84</td>
<td>0.92</td>
<td>[6]</td>
</tr>
<tr>
<td>3s$^{3}[3/2]_0 – 3p[5/2]_1$</td>
<td>0.67–0.88 33,000–36,500</td>
<td>3.3</td>
<td>0.76–0.84</td>
<td>0.92</td>
<td>[6]</td>
</tr>
<tr>
<td>3s$^{3}[1/2]_0 – 3p[3/2]_1$</td>
<td>0.67–0.88 33,000–36,500</td>
<td>3.3</td>
<td>0.76–0.84</td>
<td>0.92</td>
<td>[6]</td>
</tr>
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<td>3s$^{3}[3/2]_0 – 3p[5/2]_1$</td>
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<td>[6]</td>
</tr>
</tbody>
</table>

### Table 2
The Ne I Stark shifts $d_s$ in cm$^{-1}$ normalized to $n_e = 10^{23} \text{ m}^{-3}$

<table>
<thead>
<tr>
<th>Transition, $\lambda$ (nm)</th>
<th>$n_e$ ($10^{23} \text{ m}^{-3}$)</th>
<th>$T_e$ (K)</th>
<th>$d_s$ (cm$^{-1}$)</th>
<th>$d_s/d_{\text{in}}$</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
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<td>0.67–0.88 33,000–36,500</td>
<td>3.3</td>
<td>-0.447 (±1.6)</td>
<td>1.06</td>
<td>Tw</td>
</tr>
<tr>
<td>3s$^{3}[3/2]_0 – 3p[5/2]_1$</td>
<td>0.67–0.88 33,000–36,500</td>
<td>3.3</td>
<td>-0.433 (±1.8)</td>
<td>1.06</td>
<td>Tw</td>
</tr>
<tr>
<td>3s$^{3}[1/2]_0 – 3p[3/2]_1$</td>
<td>0.67–0.88 33,000–36,500</td>
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</tr>
</tbody>
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This table includes the range of $n_e$ and $T_e$ in the experiment. The numbers in the round brackets indicate the percentage uncertainty. A comparison with calculated values $w_{\text{in}}$ after [11] is given. Tw refers to the results of the present work.
with the theoretical values ($w_{th}$ and $d_{th}$) calculated using Eqs. (226) and (227) from [11] at the experimentally measured electron densities and temperatures. These calculations include the electron impact broadening as well as ionic corrections.

In the case of Stark widths, the best agreement is with the experimental data of [6] which were obtained in the similar range of $T_e$. The widths given in [3] exceed the results of this work by a factor of 1.3–2.0 despite similar plasma conditions. As it is stated in [3], the self-absorption might be responsible for such big broadenings. On the other hand, the discrepancy with the data presented in [10] can be explained by $T_e$ dependence of the Stark widths since they were obtained at $T_e$ about twice as high. In general, the comparison of Stark widths determined in this work with the theoretical values is within 10–20%.

For the Stark shifts, good agreement with the results of [3] is found which strongly supports the argument that the extra broadening of their lines is due to the self-absorption effect. As a rule, the lower values of the shifts are reported in [6]. The theoretical values of Stark shifts are always lower than the data of our work by a factor of 1.06–1.6.

Acknowledgments

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