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Method to measure the electric field vector in an argon glow discharge using laser polarization spectroscopy

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A method for measurement of the direction of the electric field in a glow discharge is reported. This method uses the dependence of the electronic excitation spectrum of argon atoms on the polarization of the laser radiation. In this research, laser radiation was used to excite argon atoms in a plasma from the $4s\,[\frac{3}{2}]_2$ metastable level to Rydberg levels, and excitation spectra were measured using laser optogalvanic (LOG) spectroscopy. In addition, LOG spectra of argon atoms interacting with an electric field were calculated by solving the Schrödinger equation. Good agreement was found between experimental and theoretical LOG spectra obtained for different polarizations of the laser radiation.

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The electric field is one of the most important parameters for modeling plasma discharges. Several different methods for measurement of electric field magnitudes have been reported, based on Stark spectroscopy of gaseous species such as hydrogen and helium [1–8]. The other important parameter of the electric field in the discharge plasma is the direction of the vector. For the laser-aided measurements of the direction of the electric field **F**, a polarization technique based on the dependence of the laser optogalvanic (LOG), or laser-induced fluorescence spectra on the angle between the vector **F** and the unit vector of polarization of the laser radiation $\mathbf{e}_L$ can be used. Such techniques have been reported for helium [9] and hydrogen [10]. In previous papers, we reported the development of a laser spectroscopic method for measurement of electric field *magnitude*, which used the effect of the Stark splitting of the Rydberg energy levels of argon atoms [6–8]. This method involves measurements of excitation spectra of argon atoms, and in order to populate the argon Rydberg levels, we used single-photon laser excitation from the metastable state $4s[\frac{3}{2}]_2$.

In the present paper, we report a technique to measure the *direction* of the electric field in a discharge plasma. The purpose of this paper is to describe the theory on which this measurement method is based and to demonstrate that results predicted by the theory are in good agreement with experimental results.

The spectral scheme used for this calculation and the experiment is similar to that described in [8]. Excitation is from the argon lower metastable level $4s[\frac{3}{2}]_2$ to Rydberg levels of the principal quantum number $n=9$ of argon atoms. For Rydberg states of an argon atom, the (jl) coupling scheme can be applied [11]. In this scheme, the strong coupling of the orbital momentum of the Rydberg electron **l** to the ion-core total angular momentum $\mathbf{j}_C$ results in the angular momentum of $\mathbf{K}=\mathbf{j}_C+\mathbf{l}$. The angular momentum **K** is weakly coupled to the Rydberg electron spin **s**. The total angular momentum of the whole argon atom is $\mathbf{J}=\mathbf{K}+\mathbf{s}$. Therefore, in our consideration of the Stark effect of argon atoms, we use the basis wave functions of energy levels of argon atoms in the form $\varphi=|nl[K]_J\rangle$.

We determine the positions of Rydberg energy levels and corresponding wave functions for argon atoms in the electric field **F** by solving numerically the Schrödinger equation (as explained in [8]). Since the initial state $4s[\frac{3}{2}]_2$ has a small admixture of the state $3d[\frac{3}{2}]_2$, laser excitation can populate nf Rydberg states of argon atoms. It follows from the selection rules ($\Delta K=0,\ \pm 1,\ \Delta J=0,\ \pm 1$) that within nf Rydberg levels having the same n, only two fine-structure doublets $nf[\frac{3}{2}]_{1,2}$ and $nf[\frac{5}{2}]_{2,3}$ can be populated by laser radiation in the absence of an electric field **F**. When $\mathbf{F}\neq 0$, the wave functions of the levels $nl[K]_J$ having the same n are intermixed. This leads to the appearance of a large number of Stark components in the LOG spectrum of argon atoms in an electric field. The intensity of the LOG signal from the Stark sublevel μ of the Rydberg level with principal quantum number n is proportional to the total population of this sublevel. Hence the expression for this intensity can be written as

$$I_\mu(F)=\tilde{P}_L\sum_{M=-2}^{+2}\sum_{M'}\left|\langle 3d[\tfrac{3}{2}]_2,M|re_L|\psi_\mu^{(n,M')}(F)\rangle\right|^2,$$

$$\mu=1,2\ldots,8n-24, \tag{1}$$

where the wave function $\psi_\mu^{(n,M)}(F)$ is the solution of the Schrödinger equation for the argon atom interacting with the electric field **F**, M is the quantum number of the projection of **J** onto the axis z, and the coefficient $\tilde{P}_L$ is proportional to the intensity of the laser radiation. We note that the direction of the axis z of the coordinate system $Oxyz$ is chosen parallel to the direction of the vector **F**. We assume that the laser radiation does not saturate the transitions from the level of $4s[\frac{3}{2}]_2$

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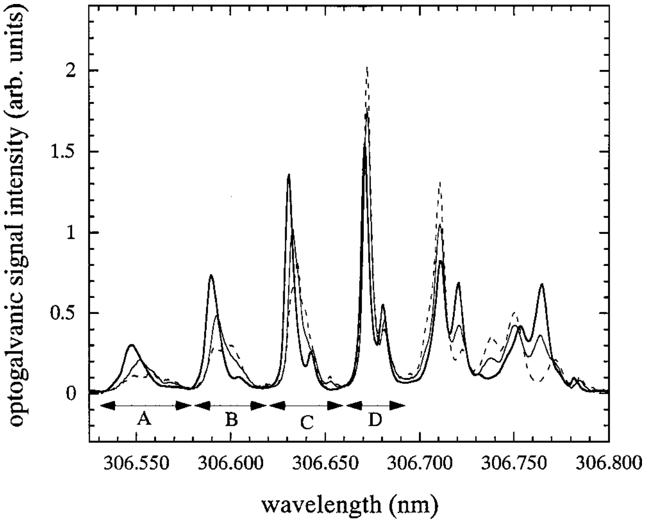

FIG. 1. Experimental LOG spectra obtained at the distance of 0.1 mm from the cathode in an argon discharge at 5.0 Torr for three angles ϑ between the vector of polarization of the laser radiation $\mathbf{e}_L$ and the discharge axis. The thick solid line corresponds to $\vartheta=0$, the thin solid line corresponds to $\vartheta=\pi/4$, and the dashed line corresponds to $\vartheta=\pi/2$. The letters *A*, *B*, *C*, and *D* show the spectral intervals that were used while integrating the profiles of the Stark components in order to determine the intensities of these components.

to Stark sublevels of the Rydberg energy level, and that the Zeeman states (with $M=0,\pm 1,\pm 2$) of the lower level $4s[\frac{3}{2}]_2$ are populated equally.

Let the vector **F** lie in the plane $x'y'$ of the laboratory coordinate system $Ox'y'x'$, but with unknown direction. Our aim is to determine this direction. It is convenient to assume that the axis x of the coordinate system $Oxyz$ lies in the plane $x'z'$ of the coordinate system $Ox'y'z'$. Let $\mathbf{e}_x$ and $\mathbf{e}_z$ be the unit vectors along the axes x and z and $\mathbf{e}_{x'}$ and $\mathbf{e}_{z'}$, be the unit vectors along the axis x' and z'. Then vectors $\mathbf{e}_{x'}$ and $\mathbf{e}_{z'}$ can be represented in the form

$$\mathbf{e}_{x'}=\cos\vartheta\mathbf{e}_x-\sin\vartheta\mathbf{e}_z, \quad \mathbf{e}_{z'}=\sin\vartheta\mathbf{e}_x+\cos\vartheta\mathbf{e}_z, \qquad (2)$$

where ϑ is the angle between the direction of the vector **F** and the axis z.

Let us consider the ratio $R_\mu(F)$ of the values of $I_\mu(F)$ calculated by using Eq. (1) for two orthogonal directions of the polarization vector $\mathbf{e}_L$ of the laser radiation, the first one being along the axis x', and the second one being along the axis z'. Substituting from Eq. (2) into Eq. (1), we obtain

$$R_\mu(F)=\frac{I_\mu^{(x')}(F)}{I_\mu^{(z')}(F)}=\frac{1+[a_\mu^{(z)}(F)/a_\mu^{(x)}(F)]\tan^2\vartheta}{\tan^2\vartheta+a_\mu^{(z)}F/a_\mu^{(x)}F}, \qquad (3)$$

where

$$I_\mu^{(x')}(F)=P_L[a_\mu^{(x)}(F)\cos^2\vartheta+a_\mu^{(z)}(F)\sin^2\vartheta],$$

$$I_\mu^{(z')}(F)=P_L[a_\mu{}^{(x)}(F)\sin^2\vartheta+a_\mu^{(z)}(F)\cos^2\vartheta],$$

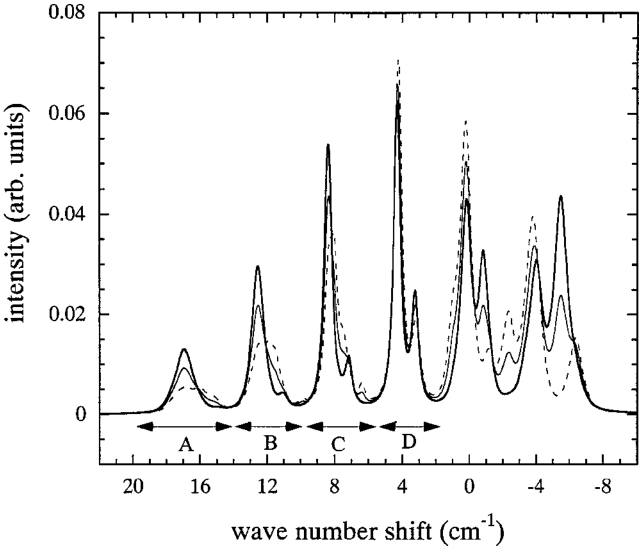

FIG. 2. Theoretical spectra of absorption of laser radiation at the transitions between lower level $4s$ $[3/2]_2$ and upper levels of $n=9$ of argon atoms calculated for the magnitude of the electric field $F=300$ V/mm and three angles ϑ between the vector of polarization of the laser radiation $\mathbf{e}_L$ and the discharge axis. The thick solid line corresponds to $\vartheta=0$, the thin solid line corresponds to $\vartheta=\pi/4$, and the dashed line corresponds to $\vartheta=\pi/2$. The letters *A*, *B*, *C*, and *D* indicate the same interval shown in Fig. 1.

$$a_\mu^{(\kappa)}(F)=\sum_{M=-2}^{+2}\sum_{M'}|\langle 3d[\tfrac{3}{2}]_2,M|\kappa|\psi_\mu^{(n,M')}\rangle|^2.$$

$$\kappa=x,z. \qquad (4)$$

Equations (3) and (4) can be used as the basis of a method to determine the direction of the electric field vector **F** in a discharge plasma. This method involves measuring the ratio of intensities of the LOG signal from the Stark sublevels μ of the Rydberg argon level for two orthogonal vectors of polarization of the laser radiation [i.e., by measuring the ratio $R_\mu(F)$ in Eq. (3)]. The angle ϑ, and hence, the direction of the electric-field vector F, can be found from Eq. (3). For this calculation, it is necessary to know the dependence of the ratio $a_\mu^{(z)}/a_\mu^{(x)}$ on F, but this can be calculated theoretically. An example of the procedure to determine ϑ from $R_\mu(F)$ is given below.

The spectroscopic scheme of the measurements and the experimental apparatus are described in detail in Refs. [6] and [7]. The laser source was a tunable dye laser pumped by a xenon chloride excimer laser. The laser was operated at λ~600 nm and the laser output was frequently doubled to generate the radiation at λ~300 nm that was needed for the experiment. The beam was directed through the plasma parallel to the electrode surfaces. Figure 1 shows experimental LOG spectra obtained from a discharge plasma at pressure of 5.0 ± 0.2 Torr in the vicinity of the cathode for the cases of three angles ϑ between the vector of polarization of the laser radiation $\mathbf{e}_L$ and the discharge axis: $\vartheta=0$, $\vartheta=\pi/4$, and $\vartheta=\pi/2$. In this experiment, the direction of the discharge electric field **F** is known to be parallel to the discharge axis. The

TABLE I. Comparison of experimental values of the ratio $[S_\eta^{(\vartheta)}]_{ex}/[S_\eta^{(\pi/2)}]_{ex}$ with theoretical values $[S_\eta^{(\vartheta)}]_{th}/[S_\eta^{(\pi/2)}]_{th}$ for each h ($h=A$, B, C and D) and for angles $\vartheta=0$ and $\pi/4$.

η	$[S_\eta^{(0)}]_{\rm ex}/[S_\eta^{(\pi/2)}]_{\rm ex}$	$[S_\eta^{(0)}]_{\rm th}/[S_\eta^{(\pi/2)}]_{\rm th}$	$[S_\eta^{(\pi/4)}]_{\rm ex}/[S_\eta^{(\pi/2)}]_{\rm ex}$	$[S_\eta^{(\pi/4)}]_{\rm th}/[S_\eta^{(\pi/2)}]_{\rm th}$
A	1.39	1.38	1.19	1.19
B	1.16	1.16	1.08	1.08
C	0.96	0.97	0.98	0.98
D	0.86	0.88	0.93	0.93

electric field magnitude is large in the region close to the cathode (the sheath region) and falls away to zero at large distances from the electrode surface. The experimental LOG spectra, presented in Fig. 1, correspond to the transitions from the lower-level $4s[\frac{3}{2}]_2$, to the upper levels of principal quantum number $n=9$. The spectra shown in Fig. 1 were normalized so that the intensity, integrated over the wavelength, is the same for each spectrum.

There are several important features of the experimental LOG spectra presented in Fig. 1. First, the spectra consist of many lines, and the profiles of the outer spectral components are broader than those of the central components. These features are directly due to the electric field, and are described in detail in Ref. [8]. By using the method described in that paper, the magnitude of the electric field was determined from the positions of the peaks in the experimental spectra to be 300±20 V/mm.

Another feature of the experimental LOG spectra is that the intensities of the spectral components are different for different values of the angle ϑ. In order to obtain the intensity of the spectral component, it is necessary to integrate the profile of the spectral component over the spectral range corresponding to this component. In Fig. 1, separate spectral ranges are designated by letters *A*, *B*, *C*, and *D*. These ranges were used to perform the integration that was used to obtain the intensities of the spectral components. In the following discussion, $[S_\eta^{(\vartheta)}]_{\rm ex}$ is the intensity of the spectral component η (where $\eta=A$, B, C, or D) in the experimental LOG spectrum corresponding to the angle ϑ between the vector $\mathbf{e}_L$ and the vector $\mathbf{F}$, which for this case is in the direction of the discharge axis. The results for the ratios of the intensities $[S_\mu^{(\vartheta)}]_{\rm ex}/[S_\mu^{(\pi/2)}]_{\rm ex}$ for $\vartheta=0$ and for $\vartheta=\pi/4$ are presented in Table I. From these results, it can be seen that the intensity $[S_\mu^{(\pi/4)}]_{\rm ex}$ constitutes about $0.5\{[S_\mu^{(0)}]_{\rm ex}+[S_\mu^{(\pi/2)}]_{\rm ex}\}$ for each η ($\eta=A$, B, C, and D) which is in accordance with Eq. (4).

Figure 2 shows theoretical Stark spectra of absorption of the laser radiation at the transition between the lower-level $4s[\frac{3}{2}]_2$ and the upper levels corresponding to $n=9$. These spectra were calculated for three angles ϑ between the vectors $\mathbf{e}_L$ and $\mathbf{F}$ ($\vartheta=0$, $\pi/4$, and $\pi/2$) and the magnitude of the electric field $F=300$ V/mm. As in Fig. 1, the letters *A*, *B*, *C*, and *D* indicated in Fig. 2 designate the spectral intervals, over which the profiles of the individual Stark components were integrated in order to obtain their intensity. We designate the corresponding intensities of these Stark components by $[S_\eta^{(\vartheta)}]_{\rm th}$ where $\eta=A$, B, C, and D. The results for the ratios of the intensities $[S_\eta^{(\vartheta)}]_{\rm th}/[S_\eta^{(\pi/2)}]_{\rm th}$ for $\vartheta=0$ and for $\vartheta=\pi/4$ are shown in Table I.

The results shown in Table I show that the experimental

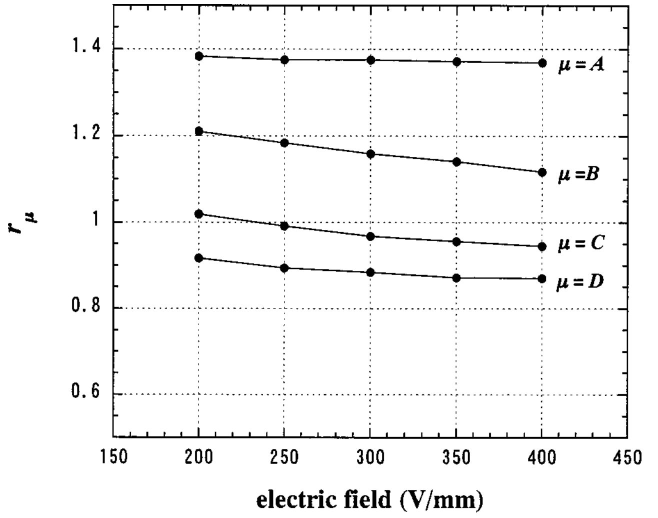

FIG. 3. The theoretical dependencies of the ratio $r_\mu=a_\mu^{(z)}/a_\mu^{(x)}$ on the strength of the electric field F ($\mu=A$, B, C, D) for the Stark spectra corresponding to the laser excitation in argon from the lower level $4s[\frac{3}{2}]_2$ to the upper levels of principal quantum number $n=9$. The letters *A*, *B*, *C*, and *D* designate the same spectral ranges as in Figs. 1 and 2.

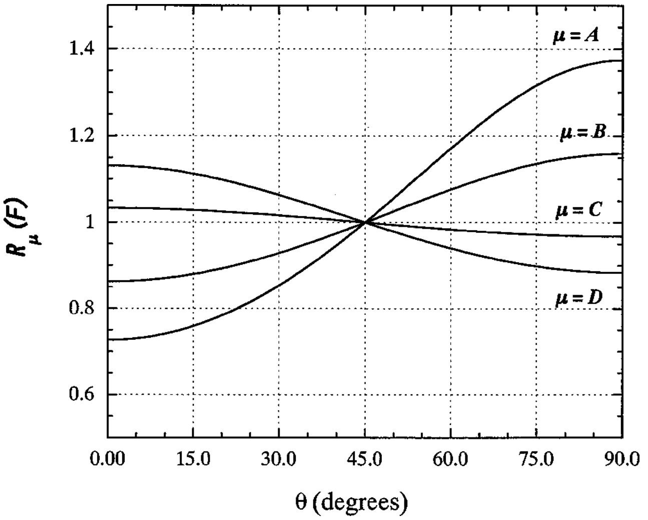

FIG. 4. The theoretical dependencies of the intensity ratio $R_\mu = I_\mu^{(x')}/I_\mu^{(z')}$ on the angle ϑ, calculated for the case of $F=300$ V/mm. The letters *A*, *B*, *C*, and *D* designate the same spectral ranges as in Figs. 1 and 2.

values of the ratio $[S_\eta^{(\vartheta)}]_{\mathrm{ex}}/[S_\eta^{(\pi/2)}]_{\mathrm{ex}}$ are consistent with the theoretical values of $[S_\nu^{(\vartheta)}]_{\mathrm{th}}/[S_\eta^{(\pi/2)}]_{\mathrm{th}}$ for each η ($\eta = A$, B, C, and D) and for both angles ϑ ($\vartheta = 0$ and $\pi/4$). This indicates that the intensities of the Stark components in the LOG spectra or argon can be used as the basis of a method to measure the direction of the electric field in a discharge plasma.

The method is based on measuring excitation spectra with the laser radiation polarized linearly in perpendicular directions, and can be explained simply as follows. The magnitude of the electric-field vector F first is determined from the measured spectra, using the relative positions of the spectral components [8]. The value of $[S_\eta^{(0)}]_{\mathrm{th}}/[S_\eta^{(\pi/2)}]_{\mathrm{th}}$ then is calculated for this value of F. Note that this ratio has the same dependence on F as the ratio $a_\mu^{(z)}/a_\mu^{(x)}$ (for $\mu = \eta$). In addition, the intensities $I_\mu^{(x')}$ and $I_\mu^{(z')}$ are obtained from the measured spectra and the ratios $R_\mu = I_\mu^{(x')}/I_\mu^{(z')}$ are calculated. The value of ϑ then can be determined by substituting R_μ and ${a_\mu}^{(z)}/{a_\mu}^{(x)}$ in Eq. (3).

Figure 3 shows the theoretical dependence of the ratio $r_\mu = a_\mu^{(z)}/a_\mu^{(x)}$ on F for $\mu = A$, B, C, and D (for the transitions $4s[\frac{3}{2}]_2 \rightarrow n = 9$). The functions $r_\mu(F)$ shown in Fig. 3 can be used together with Eq. (3) to determine the angle ϑ.

An example of this is given by Fig. 4, which shows the dependence of the ratio $R_\mu = I_\mu^{(x')}/I_\mu^{(z')}$ on the angle ϑ for the case of $F = 300$ V/mm, determined from Eq. (3) using the data shown in Fig. 3. In a practical experiment, in which the direction of the electric field was unknown, values of $R_\mu = I_\mu^{(x')}/I_\mu^{(z')}$ determined from experimentally obtained spectra could be used with this graph to determine ϑ.

The accuracy of determining the angle ϑ is mainly determined from the accuracy in measuring the intensity ratio R_μ. The size of the uncertainty in R_μ obviously depends on the quality of the measured spectrum, but a value of $\pm 10\%$ should be attainable for most conditions. By using the peak designated by A, which has the strongest dependence on ϑ, the uncertainty in ϑ should be about $15° - 20°$.

In this paper, we described the effect of laser polarization and electric field on LOG spectra obtained from argon atoms in a glow discharge. From this effect, we proposed a method to measure the direction of the electric field. The method is based on using the dependence of the LOG spectra of argon atoms corresponding to the transitions from the metastable level $4s[\frac{3}{2}]_2$ to Rydberg levels of argon atoms on the angle ϑ between the vector of polarization of the laser radiation and the vector of the electric field. Good agreement between experimental and theoretical LOG spectra of argon atoms was obtained for different angles ϑ and different spectral components.

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