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Profile of Ionized-Calcium Lines in an Arc-Plasma Jet

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Profiles of the Ca II resonance lines (λ3934 and λ3968) radiated from an arc-plasma jet were observed by means of a Fabry-Perot etalon. The measurement was made for electron densities 4.0×10^{16} and 6.4×10^{16} cm^{-3}, and a temperature around 11 500 °K. The lines exhibited Stark-effect broadening, full width at half-maximum 0.25 ± 0.06 and 0.51 ± 0.10 cm^{-1}, respectively, which was accompanied by small violet shifts, $+0.03\pm0.02$ and $+0.10\pm0.03$ cm^{-1}. The result can be interpreted by taking into account the effect of both strong and weak collisions of electrons disturbing the radiating process of the ion; the broadening is mainly contributed by strong collisions, while the shift can be explained to be due to weak collisions.

I. INTRODUCTION

RECENTLY developed theories for the Stark effect on spectral lines radiated from plasmas[1,2] have been compared by several authors with experimental results for hydrogen,[3] helium,[4] oxygen,[5] and cesium.[6] Agreement was generally good for line widths. However, Berg *et al.*[7] reported that the line He II λ4686 shifted to the blue, whereas a red shift was expected according to the Stark-effect theory; the behavior of the line was interpreted to be due to an energy shift caused by what they called plasma polarization.

The present work serves as a further test of the line-broadening theories using an ion with a simple electronic configuration. The result of an experimental study on the profile of the Ca II resonance doublet, λ3934 $(4s^2S_{1/2}-4p^2P_{3/2})$ and λ3968 $(4s^2S_{1/2}-4p^2P_{1/2})$, is presented and comparison with theory is made.

II. EXPERIMENTAL PROCEDURE

Calcium II lines were excited by means of an arc-jet excitation source, which was described in a previous work.[8] Either argon or argon-helium composite gas was passed through an electric arc burning in a nozzle-shaped electrode to form a plasma flame, into which an aqueous solution of calcium chloride (0.02%) was sprayed by means of an atomizer. A weak magnetic field was applied transverse to the arc in order to keep the arc column steady on the nozzle axis by counter-balancing the solution stream. The operating conditions are shown in Table I.

The plasma flame was approximately conical in shape, 4.5 mm in diameter at the bottom end and about 20 mm in length. The flame was ejected vertically in the upward direction, and the optical measurement was made side-on to the flame. When observed with high-speed cameras and a photomultiplier tube, the flame was found to have a conical envelope, and the emission intensity was stable except for $\pm12\%$ ripples arising from incomplete filtering in the rectifier power source.

The flame was set on the optical axis of a spectroscopic instrument, and light emitted from the center of the bottom-end portion of the flame was allowed to enter the optical system. The shift and width of the Ca II line were measured by means of a Fabry-Perot etalon, which consisted of a pair of aluminized plates separated by a 2.0- or 3.2-mm spacer, each plate having a reflecting power of 85% at λ3950 Å. An unperturbed spectrum was obtained by means of a water-cooled, hollow-cathode discharge tube. The plasma flame could be assumed to be optically thin for the H_β and Ca II lines, and for the continuum around these lines.[9]

III. DETERMINATION OF TEMPERATURE AND ELECTRON DENSITY

Since the profile measurement was made without spatial resolution, the flame not being a homogeneous light source, it was necessary to define an average temperature $\langle T(r)\rangle$ and electron density $\langle N_e(r)\rangle$, which were adequate to adopt as the basic parameters characterizing the plasma light source, r denoting the distance from the flame axis. The flame was first

TABLE I. Operating conditions of the arc-jet excitation source.

Arc current	500 A
Flow rate of plasma-forming gas	19 liter/min (Ar 42%, He 58%) 16 liter/min (Ar)
Solution-feeding rate	1.3 cc/min

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[2] H. R. Griem, *Plasma Spectroscopy* (McGraw-Hill Book Company, New York, 1964).

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[6] P. M. Stone and L. Agnew, Phys. Rev. **127**, 1157 (1962).

[7] H. F. Berg, A. W. Ali, R. Lincke, and H. R. Griem, Phys. Rev. **125**, 199 (1962).

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[9] The absolute intensity of the Ca II lines was measured and compared with the calculated intensity of a black body at the temperature of the plasma (11 500°K). An optical depth of 0.02 ± 0.01 was obtained for Ca II λ3934, which is negligible within the experimental accuracy. This method is described in Ref. 5.

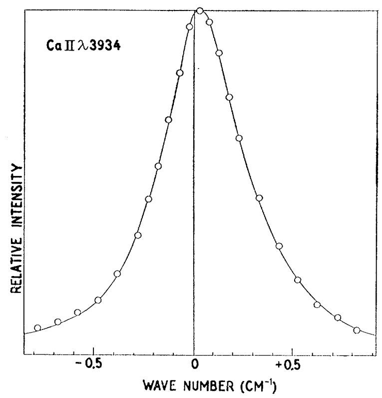

FIG. 1. Profile of Ca II λ3934 Å in Ar-He plasma.

scanned in the radial direction to obtain the spatial distribution of temperature and electron density, which were then averaged over the flame radius by including the radial distribution of the Ca II-line intensity as a weight function.

$T(r)$ was determined by comparing the measured absolute intensity of the H_β line with the theoretical expression. The positive crater of a carbon arc was used as a radiation standard,[10] and Abel's integral equation was solved numerically[11] to convert the projected intensity distribution into the true radial distribution. $N_e(r)$ was determined in the following way: The radial distribution of the relative intensity of the recombination and bremsstrahlung continuum was measured, which was approximately proportional to $[N_e(r)]^2$ according to the Kramers-Unsöld theory.[12] Absolute values of $N_e(r)$ were determined in such a way that the observed H_β profile could be fitted to the theoretical profile obtained by superposition of the profiles proposed by Griem.[13] The measured temperature and electron density are shown in Table II.[14] The electron density determined in this way agreed within 20% with that obtained by inserting $\langle T(r)\rangle$ into Saha's formula for thermal ionization.

TABLE II. Temperature and electron density in the plasma.

	Plasma-forming gas Ar-He	Ar
Average temperature (°K)	11 400±500	11 600±500
Average electron density (cm^{-3})	$(4.0\pm0.6)\times10^{16}$	$(6.4\pm1.0)\times10^{16}$

[10] M. R. Null and W. W. Lozier, J. Opt. Soc. Am. **52**, 1156 (1962).
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[12] W. Finkelnburg and T. Peters, in *Handbuch der Physik*, edited by S. Flügge (Springer Verlag, Berlin, 1957), Vol. 28.
[13] A. C. Kolb and H. R. Griem, in *Atomic and Molecnlar Processes*, edited by D. R. Bates (Academic Press Inc., New York, 1962), Chap. 5.
[14] The values on the flame axis deviate from the average by +4% for T and +25% for N_e.

TABLE III. Measured shift and width (cm^{-1}) of Ca II λ3934.

	Plasma-forming gas Ar-He	Ar
Shift	+0.03±0.02	+0.10±0.03
Full half-width		
Observed width	0.50±0.06	0.75±0.10
Instrumental width	0.08	0.13
True width	0.46±0.06	0.69±0.10
Doppler width	0.31	0.31
Stark width	0.25±0.06	0.51±0.10

IV. OBSERVED PROFILE

The observed profiles of the line Ca II λ3934 in the two cases are shown in Figs. 1 and 2. The unperturbed line is at the origin. The line is shifted to the violet and exhibits slightly asymmetric broadening. The profiles shown here must be corrected for apparent broadening due to the etalon, by subtracting half the instrumental width from the observed width.[15] The profiles were further corrected for Doppler broadening according to the folding procedure.[16] (It was assumed in these corrections that the observed profile could be approximated by a Voigt profile.) After these corrections were made, shift and broadening due to the Stark effect alone were obtained, as shown in Table III. The profile obtained for Ca II λ3968 was the same as that for λ3934 within experimental error, although some minor difference might be expected theoretically.

V. INTERPRETATION OF THE RESULTS

A violet shift of ionic lines is predicted by the theory put forward by Berg *et al.*[7] (the plasma polarization theory). However, the shift calculated according to this theory [Eq. (4–97) of Ref. 2] is 6 to 12 times as large as those actually observed, if in the calculation the lower state interaction is also taken into consideration and the

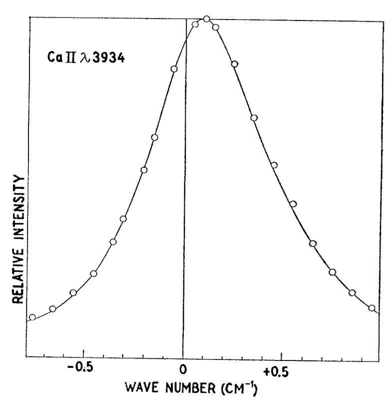

FIG. 2. Profile of Ca II λ3934 Å in Ar plasma.

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effective principal quantum numbers (n^*) are used to estimate the radial matrix elements. On the other hand, no line broadening is predicted by this theory. It therefore seems difficult to interpret the observed profiles solely in terms of the plasma-polarization effect.

Among the plasma particles disturbing the radiating process of calcium ions, only the contribution of other ions and electrons is considered here, and the role of neutral atoms as perturbers is neglected because of their short-range interaction with other atoms and ions. (However, see also Refs. 17 and 18.) We first examine the quasi-static Stark effect caused by slow ions. The level shift $\Delta\nu$ (cm^{-1}) in a static electric field F is given by

$$\Delta\nu=\gamma F^2\,, \tag{1}$$

where the Stark-effect coefficient γ can be calculated. For the Ca II resonance lines we have[19] $\gamma=+4.7\times10^{-7}$ cm^{-1}/(kV/cm)^2. Inserting the normal field strength corresponding to the measured ion density ($\approx N_e$), we obtain from Eq. (1) the calculated shift $+0.001$ cm^{-1}, which is too small to account for the observed shift. When estimated in the collision theory, the ion effect is seen to be small compared with the electron effect. Therefore, only electron collisions give appreciable contributions to the line broadening and shift.

According to the recently developed theories,[1,2,4] the effect of electron collisions is classified into strong- and weak-collision parts depending on the impact parameter ρ. In our case these theories predict line shifts which are of the same order of magnitude as the observed shifts.

The strong-collision cutoff obtained from Eq. (4–77) of Ref. 2, assuming straight perturber trajectories, is about 3 Å. Inserting this into the weak-collision part of Eq. (4–79) of Ref. 2, one gets full half-widths of 0.04 (Ar-He) and 0.07 cm^{-1} (Ar). The remaining part of the observed broadening, 0.21±0.06 (Ar-He) and 0.44±0.10 cm^{-1} (Ar), must be due to strong collisions. The full half-width for strong collisions is given by the Lorentz-Weisskopf formula:

$$w_s=\rho_{\min}{}^2N_e\langle v\rangle/c\ \mathrm{cm}^{-1}. \tag{2}$$

With $\rho_{\min}=3$ Å, this formula predicts widths of 0.09 (Ar-He) and 0.14 cm^{-1} (Ar), which are, respectively, less than half the observed widths (strong-collision part).

The cutoff deduced by an elementary consideration (see, e.g., Ref. 2)[20]

$$\rho_{\min}=e^2a_0n^{*2}/(2\langle v\rangle\hbar) \tag{3}$$

is 5.4 Å in our case; then the calculated widths, 0.26 (Ar-He) and 0.42 cm^{-1} (Ar), are in better agreement with the observed Stark widths (strong-collision part). The greater part of the Stark width is thus contributed by strong collisions, while the shift can be interpreted to be due to weak collisions.

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[20] See footnote 11 of the article of K. Murakawa [preceding paper, Phys. Rev. **146**, 135 (1966)].