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INTERNATIONAL ATOMIC ENERGY AGENCY

UNITED NATIONS EDUCATIONAL, SCIENTIFIC AND CULTURAL ORGANIZATION

1981 MIRAMARE-TRIESTE

IC/81/184

International Atomic Energy Agency
and
United Nations Educational Scientific and Cultural Organization

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STARK BROADENING OF ISOLATED U.V.-LINES OF LiI, AlI AND SiI BY A D.C.-ARC PLASMA WITHOUT AND WITH Ca *

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MIRAMARE - TRIESTE
September 1981

* Submitted for publication.

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Abstract

The halfwidths of isolated u.v.-lines of LiI, AlI, and SiI have been measured in a common d.c.-arc. Collisional broadening by neutral perturbers, electron and ion Stark effects have been calculated, without and with addition of Cs, where the electron densities were 1.64 $\times 10^{14}$ cm^{-3} and 3.54 $\times 10^{14}$ cm^{-3}, respectively, and the temperature was about 5400 K. The calculated Stark widths were compared with the relevant measured values. The errors ranged between 8% and 37%, although they involved another sources of error. It is concluded that without Cs, Doppler and neutral collisional broadenings are mainly responsible for the spectral line broadening, where Stark widths were relatively small. While with Cs, Stark widths were dominant. Corrections by quadrupole ion impact widths were considered.

1. Introduction

Stark broadening has been widely investigated and developed by several authors.[1-4] Isolated lines (whose widths are much smaller than the separation between levels that contribute to the perturbation), were treated using the impact for electrons and the quasi-static approximation for ions.[5] Higher order effects have been incorporated into the theory.[6] An alternate treatment for isolated lines has been given by Sahal-Brechote.[7] An extensive treatment of Stark broadening including tables for Stark parameters and profiles based on semi-classical calculations were done by Griem.[8] Most

of the experimental work was carried out using different sources of plasmas, e.g., wall stabilized arcs,[9-11] shock tubes,[12,13] discharge lamps,[14] inductively coupled r.f.,[15,16] etc.[8]

This work investigates Stark widths of some isolated u.v.-lines at 3232.61 A, 2881.578 A, and 3092.713 A, for LiI, AlI, and SiI, respectively, in the common d.c.-arc, which are usually used in spectrochemical analysis. The study is done under the normal conditions without and with addition of 2 ppm of Cs. The excitation temperature T = 5400 $\pm$ 62 K is determined using two FeI-lines, while the electron density N_e = 1.64 $\times 10^{14}$ cm^{-3}, and 3.54 $\times 10^{14}$ cm^{-3} is determined using two FeI and FeII-lines without and with Cs, respectively. The collision widths, $\delta\lambda_c$, due to neutral perturbers are calculated. Hence, estimation of the measured Stark width, $\delta\lambda_S^m = \delta\lambda_L - \delta\lambda_c$, (where $\delta\lambda_L$ is the measured Lorentz width of the line profile) was available. Electron Stark width , $\delta\lambda_S^e$, (corrected for ion broadening and shielding parameters), and the quadrupole ion impact widths, $\delta\lambda_S^i$, are calculated. Thus, the total calculated Stark width $\delta\lambda_S^{calc} = \delta\lambda_S^e + \delta\lambda_S^i$ was obtained. The measured and calculated Srark widths are comparable, and the errors ranged between 8% and 37%, although it involved other sources of error. It is concluded that without Cs, Stark widths were relatively small, while with Cs they were dominant. Quadrupole ion impact widths were quite important.

2. Determination of the plasma parameters

The LTE state is generally assumed to exist in the central portions of the d.c.-arc plasmas at atmospheric pressure. If this assumption is accepted, spectroscopic techniques may be combined with the Boltzman energy distribution and the Saha-Eggert ionization equilibrium relationships to yield temperature and electron density distributions.[17]

The excitation temperature T may be determined from the ratio of the intensities of two optically thin lines using the following relation

$$T = \frac{5040(E_1 - E_2)}{\log(\frac{g_1A_1}{g_2A_2}) - \log(\frac{\lambda_1}{\lambda_2}) - \log(\frac{I_2}{I_1})} \quad (1)$$

The electron density N_e may be calculated from the intensities of the emission lines of the neutral atom (^{o}), and the first ionized species ($^+$) viz.

$$N_e = 4.83\times10^{15} \frac{I^o}{I^+}\frac{g^+A^+}{g^oA^o}\frac{\lambda^o}{\lambda^+} T^{3/2} \exp(\frac{E^+ - E^o - E_i^o + \Delta E_i^o}{kT}), \quad (2)$$

ΔE_i is the lowering of ionization energy of neutral atoms by the charged particles. The notations of Eqs.(1), and (2) have their usual significance.

3.Pressure broadening

Aside from Doppler broadening, the main contribution to the spectral line broadening at the full width at half-maximum intensity (FWHM) in the d.c.-arc plasmas is from pressure effects. Pressure broadening may be caused by collisions with neutral perturbers, and/or with charged particles. Charged particle collisions produce Stark effect. However, all types of pressure broadening follow a Lorentzian distribution.

3.1 Collisional broadening by neutrals

In view of Linholm's impact theory, and assuming v.d.W-interactions between emitter and neutral perturbers (dipole-dipole

interactions), the halfwidth $\delta\nu_c$ may be given by[18]

$$\delta\nu_c = 2.71\ C_6^{2/5}\ \bar{v}^{3/5}\ N_p \ , \quad (3)$$

where, C_6 designates the difference of the v.d.W constants in the upper and lower energy states of the atomic transitions, $\bar{v}=(8kT/\pi\mu)^{\frac{1}{2}}$ is the mean value of the relative velocity of the emitter and perturber, μ is the reduced mass, and N_p is the perturber density cm^{-3}. Values of C_6 can be calculated using a Coulomb approximation by some relations stated elsewhere.[19] These relations are valid as long as the mean emitter-perturber internuclear distance $r < \rho_W$, where ρ_W is the Weisskopf radius which represents the lower limit for strong interactions.[8] It may be given by

$$\rho_W = (\ 3\pi/8\ C_6/\bar{v}\ )^{1/5} \ . \quad (4)$$

If $r > \rho_W$, Coulomb and exchange interactions should be involved (which may be caused by overlapping electrons), and hence, repulsion forces must be taken into consideration as well. In this case, the true potential function would be represented approximately by the Lennard-Jones potential.

3.2 Stark broadening

Stark broadening is caused by electric microfields from electrons and ions surrounding the emitting atoms or ions. The resulting Stark profiles depend almost exclusively on the electron (ion) density, and are only weak functions of the temperature. Electron collisions cause broadening mainly at the line core, i.e., at FWHM, while ions act at the line wings.[3] In general, isolated lines are broadened primarily by electron impacts, and relatively by crude corrections which may be sufficient to allow for ion effects. Such corrections may be estimated considering either the impact or quasi-static approximations. To do so in a practical way, one should keep in mind the overriding influence of electrons, especially in the line cores, and use a sufficient condition for the quasi-static approximation which may be valid if

$$w/(v_i N_i^{1/3}) \gtrsim 1 \quad , \tag{5}$$

where w is the electron impact width, $v_i = (2kT/m_i)^{\frac{1}{2}}$ is the ion velocity, m_i is the ion mass, and N_i is the ion density cm^{-3}. Reversing this inequality, the impact approximation will be valid.

According to Griem,[3,8] in view of the impact approximation, at the FWHM, $\delta\lambda_S^e$ of an isolated line is given by

$$\delta\lambda_S^e \approx 2w \left[1 + 1.75A(1 - 0.75R)\right] \quad , \tag{6}$$

where, w is proportional to N_e, the ion broadening parameter $A \leqslant 0.5$ is proportional to $N_e^{1/4}$, and R is the ratio of the mean distance between ions r_i, and the Debye radius ρ_D, which is given by

$$R = r_i/\rho_D = 6^{1/3}\,\pi^{1/6}\,(e^2/kT)^{\frac{1}{2}}\,N_e^{1/6} \quad . \tag{7}$$

The R-term in Eq.(7) is a measure of ion-ion correlations and Debye shielding. But for heavy perturbing ions, e.g., N^+, Ar^+, Cs^+, etc, corrections may be added to the FWHM of the isolated line produced by quadrupole ion impact width ($\delta\nu_S^i$ s^{-1}) which may be obtained by [Ref.8, page 98, Eq.(218b)]

$$\delta\nu_S^i = 2\,w_i \approx 2 \times 2\pi N_i \left[(n_i^{*2} - n_f^{*2})^2\, h\, a_0/2\pi Y^2 m\right] Z_i \quad , \tag{8}$$

where N_i is the ion density cm^{-3}, n_i^{*2} and n_f^{*2} are the squares of the effective principal quantum numbers of the transition dealt with, a_0 is the radius of Bohr's first orbit, Y is the ionization stage (Y=1 for neutral emitters), m is the mass of electron, and Z_i is the atomic number of the perturbing ionic species.

Another possibility, namely, ion broadening through second order dipole interactions (quadratic Stark effect), is not of much interest for isolated lines. It usually stays below Eq.(8) indicating that quadrupole interactions involving diagonal matrix elements are typically stronger than dipole interactions with nondegenerate states.

4. Experimental

4.1 Instrumentation

A 3.4-Ebert spectrograph (Jarrell-Ash) was used, with a 152x64 mm^2 grating (590 grooves/mm). The grating was fully illuminated yielding a theoretical resolving power of about 90,000. The reciprocal linear dispersion ≈ 4.9 A/mm is approximately constant in the wavelength range between 2000 A and 4000 A. An entrance slit of 10 μm width was used to limit the field of view to a small axial region of the arc plasma. The resolved spectra was directly recorded usig either a special direct profiling head attachment (model 15-820, Jarrell-Ash), or a blue sensitive photographic plate (Kodak). The spectral lines on the photographic plates were identified and scanned by comparison with a master plate using a digital comparator microphotometer (Jarrell-Ash).

4.2 Samples

Spectroscopic pure carbon powder, $LiCO_3$, Al_2O_3, SiO_2, CsCl, and Fe_2O_3 (Johnson Matthy Ltd.) were used. Carbon matrices containing 2 ppm of Li, Al, and Si, and 1 ppm of Fe were prepared separately. Another matrices were prepared containing the same concentrations of elements with addition of 2 ppm of Cs.

4.3 Measurements

Each element was arced (without and with Cs) under the optimal conditions: arcing current, 7 Amp; exposure time, 20 s; gap-width, 3 mm; a cup-shaped cathode; and an anode angle, 45^o. The value of 7 Amp exceeds the proper value (6 Amp) for the elements, but it is preferred to overcome the probable decrease of temperature of the arc plasma by Cs. Fe was used as a thermometric element. According to Eq.(1), and using the FeI-lines at 3719.94 A, and 3763.79 A, it was found that T = 5400 ± 62 K. The electron densities were determined using Eq.(2) for the FeI-line at 2522.85 A and the FeII-line at 2585.88 A. We found that $N_e = 1.64 \times 10^{14}$ cm^{-3}, and $3.54 \pm 0.2 \times 10^{14}$ cm^{-3} without and with Cs, respectively. The data used for E_1, E_2, E^+, E^o, E_i^o, g, and A were obtained by Bridges et al.[20] According to Unsöld,[21] a value of 0.05 eV (403 cm^{-1}) was substituted for ΔE_i^o.[22] Values of the intensities were obtained according to the following section.

5. Method of evaluation

The intensities of the investigated lines at 3232.610 A (Li), 2881.578 A (Al), and 3092.713 A (Si) were evaluated from their profiles. The measured intensities I_λ were corrected to be optically thin I'_λ, using the following relation[22]

$$I'_\lambda = - B_\lambda \ln \left[1 - \frac{I_\lambda}{B_\lambda} \right] \quad , \tag{9}$$

where B_λ is the Planck-Kirchhoff function. The measured intensity can be obtained most conveniently in units of B_λ (,i.e., I_λ/B_λ) from the direct photoelectric scan. B_λ may be given for each line by the plateau of the optically thick line core.[9] This could be done by a photoelectric scan for the lines recorded on the photographic plate by connecting the microphotometer with an XY-Inféléc recorder.

The corrected FWHM of the optically thin line profiles were $\delta\lambda^{thin}/\delta\lambda^{thick}$ = 0.89, 0.81, and 0.70 ± 0.02 for Li, Al, and Si, respectively.

The instrument function was determined to be 0.049A, and the Doppler widths were calculated to be 0.0646 A, 0.0314 A, and 0.0286 A for Li, Al, and Si, respectively, at the lines referred to before. Calculation of the Gaussian component $\delta\lambda_G = \left[(\delta\lambda_{inst})^2 + (\delta\lambda_D)^2\right]^{\frac{1}{2}}$ at FWHM of the line profile was then available. Thus, deconvoluting[23] the corrected FWHM, $\delta\lambda^{thin}$, the measured Lorentzian widths, $\delta\lambda_L$, were obtained, Table 3.

6. Results and discussions

The Lorentzian width, $\delta\lambda_L$, of the line at FWHM may be attributed to two Lorentzian components produced by collisions of the emitters with neutral perturbers, $\delta\lambda_c$, and with charged particles (electrons and ions), $\delta\lambda_S$, i.e., Stark width. $\delta\lambda_L$ is a sum of $\delta\lambda_c$ and $\delta\lambda_S$, since they are scalarly additive. Thus, using the measured values of $\delta\lambda_L$, the measured values of $\delta\lambda_S^m$ can be estimated once, values of $\delta\lambda_c$ were known.

6.1 Calculation of $\delta\lambda_c$

From Table 1, it is obvious that the calculated values of the Weisskopf radii ρ_W (using Eq.5), were quite larger than the internuclear distances between the emitters and perturbers (,e.g., N_2). Consequently, the repulsive forces were neglected, and the Lennard-Jones potential was excluded. However, the attractive v.d.W.-forces were considered, and then using a Coulomb approximation, values of the force constants C_6, were calculated,[18,19] (Table 2). Neutral perturbers at 1 atmospheric pressure in the d.c. arc plasmas constitute mainly of N_2 (which is dominant), O_2, and C_2.[22] Thus, using Eq.(3), values of $\delta\lambda_c$ were calculated, Table 3. These values might be considered constant approximately, without and with Cs.

6.2 Calculation of $\delta\lambda_S$

On the basis of Lindholm-Foly's impact theory, $\delta\lambda^e_S$, were calculated using Eq.(6). The electron impact widths, w, and the ion broadening parameters, A, were determined from the tables of Griem,[8] to fit the values of $N_e = 1.64 \times 10^{14}$ cm^{-3}, and T = 5400 K. Values of R were calculated by Eq.(7).

For ion broadening, the inequality given by Eq.(5) was examined. Considering that $N_i=N_e=1.64 \times 10^{14}$ cm^{-3}, it was found that $w/(v_i N_i^{1/3})$ = 0.03, 0.02, and 0.003 for Li, Al, and Si, respectively, i.e., $\ll 1$. Hence, the impact approximation was valid. The ionic species are N^+, O^+, and C^+ (N^+ are dominant).[22] The quadrupole ion impact widths, $\delta\lambda^i_S$, were calculated using Eq.(8). Values of $\delta\lambda^e_S$, and $\delta\lambda^i_S$ are reported in Table 3.

In a presence of Cs, the intensities of the spectral lines were enhanced and broadened. Such broadening might be attributed to stronger Stark effect. For an electron density $N_e = 3.54 \times 10^{14}$ cm^{-3}, values of $\delta\lambda^e_S$ were calculated using Eq.(6), as it was explained before. But for ion broadening, it should be taken into consideration that $N_i = 3.54 \times 10^{14}$ cm^{-3} is a sum of 1.64×10^{14} cm^{-3} without Cs, and 1.9×10^{14} cm^{-3} of Cs^+. The quadrupole ion impact widths, $\delta\lambda^i_S$, due to Cs^+ were calculated, (plus those attributed to another ionic species without Cs), and included also in Table 3.

6. Conclusions

From Table 3, it could be concluded that:

1) Without Cs: the spectral lines were essentially broadened by Doppler, $\delta\lambda_D$, and by collisions with neutral perturbers, $\delta\lambda_c$, while Stark widths were relatively small. Broadening by electrons was very small such that it may be neglected, while quadrupole ion broadening was remarkable. The total Stark width was $\delta\lambda^{calc}_S = \delta\lambda^e_S + \delta\lambda^i_S$.

2) With Cs: although broadenings by Doppler and neutral collisions were still considerable, Stark effect was dominant. Quadrupole ion broadening played a major role, such that its contribution was quite larger than that of either Doppler or neutral collisional broadenings. However, $\delta\lambda^{calc}_S = \delta\lambda^e_S + \delta\lambda^i_S$ ($N_i = 1.64 \times 10^{14} cm^{-3}$) + $\delta\lambda^i_S$ ($N_i = 1.9 \times 10^{14} cm^{-3}$ of Cs^+).

3) The values of calculated Stark width, $\delta\lambda^{calc}_S$, were comparable with the relevant experimental values $\delta\lambda^m_S$. The errors ranged between 8% and 37%, although they involved uncertainities associated with $\delta\lambda_L$, and $\delta\lambda_c$.

ACKNOWLEDGMENTS

The author would like to thank Professor Abdus Salam, the International Atomic Energy Agency and UNESCO for hospitality at the International Centre for Theoretical Phyiscs, Trieste.

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Table 1

Element	Li	Al	Si
ρ_W A	5.83	4.90	5.12
r A	1.45	1.25	1.10
r (N_2) = 1.75 A			

Table 1. Values of the Weisskopf radii, ρ_W, and the atomic radii, r, of Li, Al, and Si; and the molecular radius r of N_2 as a perturber.

Table 2

Element	Transition	Wavelength	n_i^{*2}	n_f^{*2}	C_6 $cm^6 s^{-1}$
Li	2S-3P	3232.610 A	2.522	8.769	2.64×10^{-31}
Al	3P-3D	3092.713 A	2.272	6.900	9.17×10^{-32}
Si	3P-4S	2881.578 A	1.670	4.460	8.50×10^{-32}

Table 2. Values of the squares of the effective principal quantum numbers of the initial, n^{*2}, and final, n_f^{*2}, states for the transitions dealt with; and the relevant force constants C_6.

Table 3

Element	LiI 3232.610 A		AlI 3092.713 A		SiI 2881.578 A	
	without Cs	with Cs	without Cs	with Cs	without Cs	with Cs
$\delta\lambda^{thick}$ A	0.1350	0.2400	0.0950	0.1450	0.1000	0.1200
$\delta\lambda^{thin}$ A	0.1200	0.2150	0.0768	0.1200	0.0700	0.0845
$\delta\lambda^m_L$ A	0.0630	0.1800	0.0298	0.0900	0.0193	0.0440
$\delta\lambda_D$ A	0.0646		0.0314		0.0286	
$\delta\lambda_c$ A	0.0416		0.0188		0.0158	
$\delta\lambda^e_S$ A	0.0030	0.0063	0.0009	0.0020	0.0002	0.0004
$\delta\lambda^i_S$ A	0.0117	0.1209	0.0062	0.0611	0.0020	0.0195
$\delta\lambda^{calc}_S$ A	0.0147	0.1272	0.0071	0.0631	0.0022	0.0199
$\delta\lambda^m_S$ A	0.0214	0.1384	0.0110	0.0712	0.0035	0.0282
Error%	31.30	8.10	35.50	11.40	37.14	29.40

Table 3. Values of the halfwidths at FWHM due to different types of broadening.

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