Stark-width measurements of singly ionized calcium resonance lines in a wall-stabilized arc

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Stark widths of the Ca II 3933.7- and 3968.5-Å resonance lines are measured around a temperature of 13 000 K and an electron density of 10^{17} cm^{-3} in a stationary wall-stabilized arc burning in argon with a small addition of Ca vapor. The electron density is measured using the Stark width of the hydrogen Hβ line. Optical-depth measurements are performed at the peaks of the investigated lines and the calcium concentration is carefully controlled to minimize the reabsorption. A Fabry-Pérot interferometer, whose instrumental profile is measured, yields the required high spectral resolution. The experimental profiles are analyzed using a least-squares fit of the Voigt function to all profile intensity points. The width of the Ca II resonance doublet is found about a factor of 2 lower than both semiclassical and full quantum calculations, in agreement with the earlier experiments of Yamamoto and of Roberts and Eckerle.

I. INTRODUCTION

Owing to the comparative simple structure of the set of perturbing levels involved, the electron impact Stark widths of the resonance lines of Mg II and Ca II ions have been calculated through various semiclassical approximations, as well as through completely quantum-mechanical treatments. Because of the computational effort, however, the latter consider only a small number of interacting states. Measurements of these line widths under well-known physical conditions are then of interest to appreciate the reliability of such calculations.

In a recent paper,[1] hereafter referred as I, we present measurements of the Mg II resonance linewidths, whose results agree very well with the full quantum calculations of Barnes[2] and lie a factor of 2 lower than most of the semiclassical calculations and other experimental results.

In the case of Ca II, the discrepancy between the semiclassical[3,4] and the quantum-mechanical[5] calculations is of the order of 20% and is small compared with the large discrepancies prevailing between previous experimental results[4,6–12] which spread over more than a factor of 2. As the typical error of these experimental width results is claimed to be of the order of $\pm 20\%$, the large spread of the width values implies the presence of unnoticed systematic errors. In order to increase the reliability of the present results, we based our measurements on experimental methods allowing a careful check of the most significant sources of systematic error: electron density determination, spectral resolution, deconvolution procedure, optical-depth effects.

II. WIDTH MEASUREMENT METHODS

All experimental technics are thoroughly described in I and they are only briefly reviewed hereafter. The plasma source is a wall-stabilized arc burning in argon (plasma length 75 mm, diameter 5 mm) at normal pressure. Pure calcium is heated in a furnace around 600 °C, and calcium vapor is carried by an argon flow into the central part of the arc column. Reverse flows of argon avoid the diffusion of calcium impurities in the end zones of the arc.

The plasma is imaged end on (magnification 1:1, beam aperture 1/120) on the entrance slit of a grating monochromator set at a fixed wavelength and acting as a predispersor. A piezoelectrically scanned Fabry-Pérot interferometer (working plate separation 1 mm) mounted behind the exit slit yields high-resolution scans of the Ca II resonance lines.

A measurement and control microprocessor is interfaced between a minicomputer and the experiment. For a given sequence of measurements, the wavelength scans of the monochromator together with the translation of the plasma source, the acquisition of the photomultiplier output, the data storage and treatment are programmed. The various experimental runs are then performed entirely automatically, which allows an enhanced repetition rate of the measurements.

The instrumental profile of the Fabry-Pérot interferometer is measured, as described in I, by decreasing the plate separation until the instrumental width becomes larger than the width of the Ca II resonance lines delivered by a hollow cathode lamp. The values of the Gauss and Lorentz widths of the instrumental function, obtained through a least-squares fit of the measured profile by a Voigt function, are reported in Table I. The main systematic error, relevant to the determination of an instrumental profile with a not strictly monochromatic line, yields a 6% overestimation of the Lorentzian width.

The optical depth at the peak of the Ca II resonance lines is measured directly by a double-path method using a concave mirror. The furnace temperature and the argon flow are adjusted to minimize the reabsorption and the only profiles considered as free of reabsorption and kept for further analysis correspond to $\tau < 0.1$ (i.e., an overestimation of the Lorentzian width due to self-absorption lower than 2.5%, see I).

All the end-on measured intensity data within a free spectral range of the Fabry-Pérot interferometer are fitted by a least-squares analysis. The fitting function is the sum

TABLE I. Instrumental profile determination. The absolute widths are given for a plate separation of 1 mm. The indicated uncertainties are random.

λ (Å)	Apparatus finesse	Reflectivity finesse	Full width of the Voigt profile (Å)	Full Lorentzian width (Å)	Full Gaussian width (Å)
3933.7	75.5	110	0.010 24±0.2%	0.007 03±0.5%	0.0057±1%
3968.5	76.4	105	0.010 31±0.3%	0.007 50±0.8%	0.0054±2%

of a constant continuum and of three Voigt profiles (in the analytical approximation of Whiting[13]) accounting for the overlapping of interference orders. The free parameters of the fit are the Voigt and Lorentz full widths, the position and intensity of the line peak, and the continuum value. The Stark width of the line l_s is then calculated through the relation $l_s = l_{\text{meas}} - l_I - l_{\text{vdW}}$, l_{meas} being the measured Lorentzian width given by the least-squares fit, and l_I the instrumental Lorentzian width. The van der Waals width l_{vdW} is calculated following Griem[14] and amounts to 10% of l_{meas} at most. The resonance broadening is insignificant due to the very low density of calcium atoms. It should be noted that, for Ca II resonance lines, the impact approximation is valid for ionic perturbers as well as for electrons,[4] leading to a Lorentzian profile. The measured Lorentzian width is then the total (electrons plus ions) Stark width.

III. PLASMA PARAMETER DETERMINATION

The plasma temperature on the arc axis is deduced from end-on measurements of the absolute emission coefficients of Ar I 4300-Å and Ar II 4806-Å lines (transition probability values from Nubbemeyer,[15] carbon arc calibration from Magdeburg and Schley,[16] systematic error $\Delta T/T = \pm 2\%$). As shown in I, axial temperature inhomogeneities induced by the plate structure of the cascade arc are completely negligible.

The electron density on the arc axis is deduced from end-on measurements of the Hβ line Stark width. As, for a given arc current, the electron density in the argon-hydrogen mixture differs from the density in the pure argon plasma and as accurate Hβ profiles require the knowledge of the argon continuum and of the photomultiplier response over a large spectral range, the following procedure (described for instance by Helbig and Nick[17]) is performed.

The emission of the pure argon plasma at a given current is measured at 700 points over the 4700—5000-Å range, together with the continuum at 5320 Å. Then the argon-hydrogen mixture (2% H_2 in argon) is introduced in the central part of the arc and the current is varied so that the continuum emission I_c at 5320 Å is the same for both Ar-H and pure Ar plasmas. The change in the plasma temperature induced by this change in the current is measured using the Ar I 4300-Å line and found to be lower than 1%. As $I_c \propto n_e^2 T^{-1/2}$ this change in temperature corresponds to an electron density variation lower than 0.25%. The equality of the continua of both Ar-H and Ar plasmas insures practically the equality of electron density values at the two different currents. The emission of the Ar-H plasma is then measured at the same points as for the pure argon plasma. A calibration run with the carbon arc at the same 700 points closes the measuring sequence. The difference between the two calibrated spectra yield the desired profile of the Hβ line.

This profile is checked for reabsorption and inhomogeneity: The optical depth of Hβ is measured by the double-path method and found to be lower than 0.02, corresponding to an insignificant overestimation of the width. The homogeneity of the Hβ emitting plasma layer is tested by measuring the ratio of peak separation to halfwidth of Hβ, sensitive to inhomogeneites (Wiese[18]). This ratio is found to be independent of the electron density and very close to the theoretical values (see Table II).

The electron density is then deduced from the width of the Hβ profile, using the tables of hydrogen Stark broadening of Vidal, Cooper, and Smith.[19] The results are given in Table II. For comparison, the electron density values deduced from continuum measurements at 4000 Å (with ξ factor equal to 1.5, Hofsaess[20]) are given. The agreement is good. This last method is used only as a routine check, for instance to verify that the electron density value is unaffected by calcium admission in the arc column.

The random error in the electron density determination (standard error of the mean value) is ±0.5% and ±1.3% for 60 and 100-A arc currents, respectively. The systemat-

TABLE II. Plasma diagnostics; end-on measurements, pressure 1020 mbar.

Arc current (A)		60	100
Temperature (K)	Ar I 4300-Å line	12 240±250	13 300±270
	Ar II 4806-Å line		13 400±270
Electron density (10^{17} cm^{-3})	Continuum 4000 Å	0.81 ±0.16	1.35 ±0.27
	Hβ halfwidth	0.80 ±0.08	1.32 ±0.13
Ratio peak separation to halfwidth of Hβ	This experiment	0.356±0.019	0.356±0.015
	Vidal *et al.* (Ref. 19)	0.37	0.37
	Kepple and Griem (Ref. 26)	0.35	0.35

TABLE III. Measured Stark widths of Ca II resonance lines.

Arc current (A)	60		100	
Temperature (K)	12 240		13 350	
Electron density (10^{17} cm^{-3})	0.80		1.32	
Neutral density (10^{17} cm^{-3})	4.51		2.97	
	3933.7 Å	3968.5 Å	3933.7 Å	3968.5 Å
van der Waals full width (Å)	0.009 56	0.009 72	0.006 52	0.006 64
Stark full width (Å)	0.0914	0.0846	0.180	0.161
Random error (%)	±2.4	±2.8	±2.4	±6.9
Systematic error (%)	±5	±5	±5	±5
Stark full width (Å) Normalized to $n_e = 10^{17}$ cm^{-3}	0.115	0.106	0.136	0.122
Random error (%)	±2.8	±3.1	±4.2	±7.7
Systematic error (%)	±15	±15	±15	±15

ic error in the measured width $\Delta\lambda_{1/2}$ of Hβ (arising from wavelength calibration and axial inhomogeneities) is at most ±2%. The theoretical values of the reduced halfwidth $\Delta\alpha_{1/2}$ calculated by Vidal, Cooper, and Smith[19] are compared in Ref. 17 with experimental results involving high-precision independent electron density determination. It appears that the experimental reduced widths agree with the calculated one within ±4% in the region of interest, and this value is taken as the systematic error in $\Delta\alpha_{1/2}$. As $n_e\alpha(\Delta\lambda_{1/2}/\Delta\alpha_{1/2})^{3/2}$, the total systematic error in n_e is about ±10%.

IV. RESULTS AND DISCUSSION

The results of the present work are given in Table III and are compared with the other calculated and measured values in Fig. 1. Details on the error assessment method can be found in I. The random error is dominated by the noise in the experimental profiles, measured by the standard deviation of the Lorentz width given by the fitting code (typically 20%). The final random error on the weighted mean is greatly reduced due to the large number of registered profiles (around 50 over three independent experiments). The systematic error on the reduced Stark width (at $n_e = 10^{17}$ cm^{-3}) comes mainly from systematic error in electron density determination.

Figure 1 shows clearly the propensity of the experimental results to gather together in two groups: Our results agree with the ones of Yamamoto[6] and Roberts and Eckerle,[7] while the other values[4,8–12] lie between 1.4 and 1.8 times higher (disregarding the rather small variation of the width with the temperature). No systematic trend in the experimental results, related either to the type of the plasma source or to the plasma diagnostic method is obvious. Nevertheless, a review of the experiments draws attention to several points: Jones *et al.*[9] do not mention any measurement or even any check of the optical depth (to bring their results in agreement with ours, an undetected optical depth around 0.8 should be assumed). Roberts and Barnard[10] claim that the Ca II lines are optically thin only side on but the electron density and calcium line emission are found uniform over 60% of the plasma diameter only and the way to take this nonuniformity into account is not stated. Yamamoto[6] and Fleurier *et al.*[4] measure also a profile emitted radially from an inhomogeneous plasma and calculate the relevant mean electron density along the diameter through different procedures, both of them involving an obvious source of experimental and computational uncertainty. It is also of interest to know that, before deconvolution, our width values are about 1.5 times larger and would be then in fairly good agreement with the other group of values.

Finally, it should be stated that the widths of both lines of the resonance doublet are generally not given, or found to be the same within the experimental uncertainty.[6,11] In contrast, we find the $J = \frac{1}{2}$-$\frac{3}{2}$ line around 10% wider than

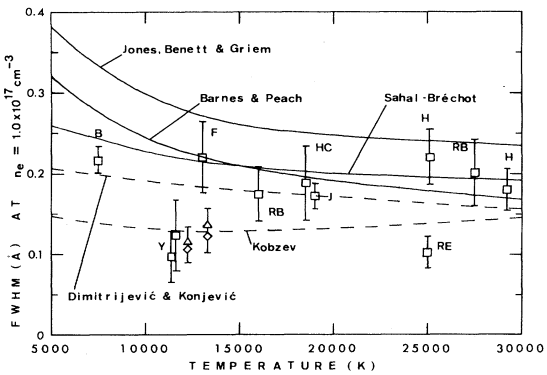

FIG. 1. Comparison of our Ca II results with published data. Y, Yamamoto (Ref. 6); RE, Roberts and Eckerle (Ref. 7); HC, Hildum and Cooper (Ref. 8); J, Jones *et al.* (Ref. 9); RB, Roberts and Barnard (Ref. 10); H, Hadžiomerspahić *et al.* (Ref. 11); F, Fleurier *et al.* (Ref. 4); B, Baur and Cooper (Ref. 12). Δ and ◇: this experiment for the $J = \frac{1}{2}$-$\frac{3}{2}$ and the $J = \frac{1}{2}$-$\frac{1}{2}$ line, respectively (the error bars represent the systematic error).

the $J=\frac{1}{2}$-$\frac{1}{2}$ line, as in the case of Mg II. Though quite small, this difference is experimentally significant for it is larger than the random uncertainty on the measured width.

As to the results of the semiclassical calculations (Jones *et al.*,[3] electron broadening; Sahal-Bréchot,[4] electron broadening including a correction for resonances of cross sections) and of full quantum calculations (Barnes and Peach,[5] electron broadening), they all lie about a factor of 2 higher than our results. The good agreement found in the case of Mg II resonance doublet with the quantum results of Barnes[2] (see I) is not met in the case of Ca II doublet with the quantum results of Barnes and Peach,[5] both calculations using the close-coupling results of Burke and Moores[21] for the scattering of electrons by Mg II or Ca II ions. Bely and Griem[22] outline that the important 4*D* level is not included in the close-coupling expansion for Ca II (but adding one more perturbing level would lead to a larger width and then to an increased discrepancy with our results). On the other hand, as outlined by Jones *et al.*,[9] the large number of energy levels near the 4*S* and 4*P* levels of the transition leads to many more resonances in the scattering cross sections. Fully quantum calculations seem to be less reliable for Ca II than for Mg II, for which such difficulties are not present.

Lastly, the agreement is better between our results and the semiempirical formula of Dimitrijević and Konjević,[23] which use an analytical expression, derived by Younger and Wiese,[24] for the effective Gaunt factor appearing in the expression of the inelastic cross sections. The agreement is very good with the semiempirical calculations of Kobzev[25] where the effective Gaunt factor is derived from a comparison with quantum-mechanical results of the excitation cross sections.

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