Stark broadening of neutral xenon

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Stark broadening of prominent visible Xe I lines are measured in three independent experiments. The three studies all use quiescent shock-tube plasmas as emission sources, but each employs distinctly different instrumentation, shock-heating regimes, and analytical methods. Owing to conditions in the light sources, temperatures between 9200 and 12 500 K, and electron densities in the range $(2.2-10.0)\times 10^{16}$ cm^{-3}, competing broadening mechanisms and radiative trapping do not seriously complicate the Stark-width determinations. The three sets of broadening results are in mutually close agreement. Generally, they also compare satisfactorily with previous measurements. For some lines, the observed broadening disagrees with theoretical predictions.

I. INTRODUCTION

Experimental contributions toward an understanding of Stark broadening in the rare gases have largely focused on helium, neon, and argon.[1,2] For Xe I some shock-tube data on Stark widths and shifts,[3,4] and calculations[4–6] of quadratic Stark constant C_4 have been published. However, earlier measurements do not converge well while use of static Stark constants for adiabatic collision widths is questionable for such lines as 4624 and 4671 Å.[3]

The traditional error proneness of width versus electron-density data was taken into account at the outset of the present work. To counter possible systematic errors the plasma state and line broadening were obtained by largely independent techniques in three separate sets of experiments. Line-broadening (semiempirical,[7] electron-impact,[8] and quasistatic[9,10]) models are used to provide first experiment-theory comparisons of Stark parameters for Xe I.

II. EXPERIMENTAL

The spectroscopic shock tubes, photometry, source diagnoses, and methods of data reduction are individually treated elsewhere.[4,11–13] The most salient features of the three parallel experiments are compared in Table I. The experiments designated 1 and 3 used the same shock tube (with a circular cross section) and observations were made in the plasma behind incident shock waves. In experiment 2 a rectangular tube was utilized and emission was observed from plasmas behind first and multiply reflected shock waves.

In experiments 1 and 3 plasma temperatures were deduced from Saha-Boltzmann relations applied to measured plasma pressures and electron densities. Temperatures in experiment 2 were found from line-reversal intensities[14] at the center of optically thick H_α, and from the measured absolute integrated intensities of Ne I $\lambda=5852$ Å and H_β. The mean of these latter temperature determinations is reliable to $\pm 5\,\%$. In all experiments plasma pressures were recorded with an accuracy of $\pm 5\,\%$ by a fast piezoelectric transducer.

Electron densities were obtained via an Ashby-Jephcott laser interferometer operating in the visible ($\lambda=6328$ Å) and infrared ($\lambda=33\,900$ Å) in experiments 1 and 3, respectively. Neutral contributions to the plasma refractive index were taken into account yielding electron-density precisions of $\pm 5\,\%$. Electron density was found in experiment 2 by fitting the symmetrized (red-blue averaged) experimental H_β profile to theoretical Stark shapes.[15,16] Depending on signal-to-noise ratios and mean spectral densities H_β widths can be determined to $10-15\,\%$ precisions, which translate into a $15-22\,\%$ random error for any single electron-density determination.

Time-resolved recording of xenon lines in all three experiments relied upon fast photographic emulsions. Pronounced grain was the limiting factor on resolution. Emulsions were calibrated with a carbon arc in the prescribed way.[17] Calculated Voigt shapes[18] were the bases for deconvolution procedures in experiments 1 and 2. In order to account for line asymmetry and to facilitate the least-squares smoothing of grain noise, Fourier-transformation deconvolution techniques were used

TABLE I. Salient features of the three independent experiments.

	Richou expt. 1	Lesage and Miller expt. 2	Truong-Bach expt. 3
Shock tube			
Section	Circular, ϕ 5 cm	Rectangular, 7×9 cm^2	Circular, ϕ 5 cm
Test gas	Xe	0.5–3 at.% Xe + (0.5–1 mol.% CH_4 or SiH_3)+Ne	40 at.% Xe in Ar + $(CH_3)_3Al$
Initial pressure	1–5 Torr	10–25 Torr	13 Torr
Driven gas	He, 60–150 bars	H_2, 60–90 bars	H_2, 170 bars
Shock waves	incident	reflected	incident
Spectrograph	prism, *F*/4.7	Czerny-Turner, *F*/7	echelle grating, *F*/5
Instrumental width	2–4 Å	0.3 Å	0.05 Å
Emulsion	Kodak recording 2475	Kodak recording 2475	Kodak recording 2485
Time resolution	drum camera	fast shutter	rotating disk
Plasma diagnostic			
Electron density	laser interferometer	H_β profile	laser interferometry
Pressure	quartz transducer	quartz transducer	quartz transducer
Temperature	Saha-Boltzmann	H_α, H_β, Ne I $\lambda=5852$	Saha-Boltzmann
Plasma parameters			
Electron density,	$(2.2-7.1)\times10^{16}$ cm^{-3}	$(6-10)\times10^{16}$ cm^{-3}	9×10^{16} cm^{-3}
Pressure, or density	calculated	measured	measured
Temperature	9200 K	10 000–12 500 K	9600 K
Instrumental	Voigt profile	Voigt profile	Fourier transformation
Doppler, van der Waals, others	negligible	negligible	negligible

in experiment 3. van der Waals, resonance, and isotope broadenings are all negligible.[9,10] Tests in plasmas behind reflected shock waves[19,20] disclosed no repeatable inhomogeneities.[21] Estimates of boundary-layer effects in experiment 3 indicate that reabsorption of visible lines is negligible.[22] This finding was applied to experiment 1 for which measured peak optical depths were less than 0.02. The absence of significant radiative trapping in experiment 2 was ascertained by comparing the brightnesses of xenon lines with brighter nearby neon lines or with a H_α line (where the line reversal directly determines the Planck intensity[23]).

Shifts of Xe I lines were measured in experiment 3 by superimposing thorium wavelength fiduciary lines on the shock-tube spectra. Wavelength displacements were read off from microdensitometer tracings or directly measured by using an optoelectronic comparator.

III. THEORETICAL

A general semiempirical formulation for electron-broadened widths has been proposed by Griem [see Eq. (33) of Ref. 7]. We calculated[24] these semiempirical widths for Xe I by replacing dipole matrix elements with calculated oscillator strengths[25] and by using effective Gaunt factors extrapolated down to zero energy.[26] However, this procedure underestimated[24] experimental widths by a factor of ~2. Similarly, in an earlier comparison of semiempirical Stark widths for neutrals Miller and Bengtson found[23] that the theory tended to underpredict by a factor of 1.5 for several elements. Omission of elastic collisions and ion effects and/or unrefined estimates of Gaunt factors for complex atoms[7,27] may be responsible for this trend.

Calculation of electron-broadening parameters can be approached through semiclassical impact theories,[8–10] predictions of neutral linewidths, and shifts in this approximation having been published for He to Cs.[9,10] For complex atomic systems the elaborated calculations can be handled by Sahal-Bréchot's computer code.[8] Calculations of the dipole part[8] of the electron-impact width and shift are feasible when atomic oscillator strengths of collision-induced transitions between upper and lower states of a line are known. For Xe I these oscillator strengths have been computed by Aymar and Coulombe.[25] Their central field model which takes into account intermediate coupling and configuration mixing has been partially verified against measured line strengths.[25] Sahal-Bréchot[28] has calculated the quadrupole-broadening contributions for atoms obeying *LS* coupling. Truong-Bach and Drawin have derived[29] analogous expressions for the rare gases whose energy levels are described by *jl* coupling.[30,31] These expressions take into account complex configurations (of type $l_1^n l_2$) outside the closed shells as appropriate for xenon. Radial matrix elements were obtained by solving the Schrödinger equation with scaled Thomas-Fermi potentials of Stewart and Rotenberg.[32]

We computed the electron-impact widths w_0 and shifts d_0 of neutral xenon for the plasma conditions of $T_0=10^4$ K and $N_{e,0}=10^{17}$ cm^{-3} according to Refs. 8 and 29. Assuming ion quasistatic broadening the total Stark widths W_0 and shifts D_0 to be compared with experiment were then calculated from the Griem's formula,[9] or by direct computations of half-intensity points of whole profiles[9] averaged by the Hooper's ion-microfield distributions.[33]

IV. RESULTS AND DISCUSSIONS

Uncertainty in measured Stark widths stems primarily from random errors in electron-density determinations and profile deconvolution. To reduce chances for systematic error arising in deconvolution, the experimental regime was made to span a wide range of electron densities (see Fig. 1). Error estimates are $\pm\sigma/\sqrt{n}$. Stated confidence limits are 90% for experiments 1 and 2. For experiment 3 ($T_0=10^4$ K and $N_{e,0}=10^{17}$ cm^{-3}) instrumental widths are negligible compared to Stark widths. Our method of deconvolution by Fourier transforms conserves the inherent asymmetry of Xe I Stark profiles and smoothes grain noise to a standard deviation of a few percent.[13] Precision for the Stark-width determination in experiment 3 is estimated to about 10%.

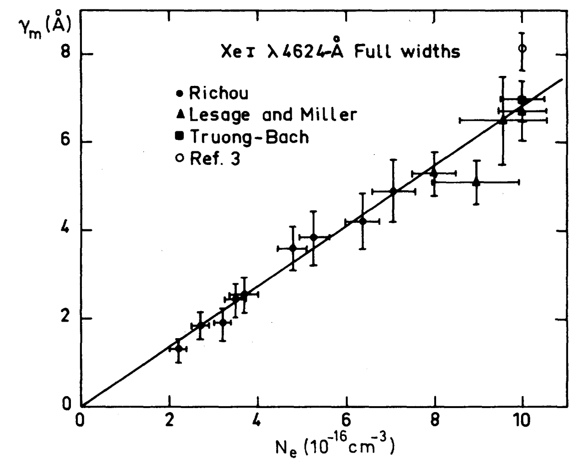

FIG. 1. Experimental full half-width γ_m versus electron density N_e for the Xe I $\lambda=4624$-Å line.

To provide a uniform basis for comparison measured Stark widths at various plasma conditions are linearly normalized to $T_0=10^4$ K and $N_{e,0}=10^{17}$ cm^{-3}. Normalized results from our three sets of experiments are given in Table II. Minor systematic differences between the three sets of values could be attributed to nonlinear temperature and electron-density dependence of the Stark broadening. With the index 0 and x referring, respectively, to the reference and actual plasma conditions, the electron-impact widths w, total Stark widths W, and electron densities N_e are interrelated by $w_e/w_x=N_{e,0}/N_{e,x}$ and

$$\frac{W_0}{W_x}=\frac{1+1.75q_0\alpha_0}{1+1.75q_x\alpha_x}\frac{N_{e,0}}{N_{e,x}} .$$

Here $q(N_e,T)$ and $\alpha(N_e)$ are, respectively, the Debye-shielding and ion-broadening parameters, and w is practically proportional to N_e for our experimental temperature range [see Eqs. (4-68) and (4-90) of Ref. 9]. If the experimental width $W_{\rm expt}$ can be compared with the total Stark width W_x we will have $W_{\rm expt}=W_x$, and the measured value $W_{\rm expt}$ when normalized to the reference conditions will be

$$W_m=\frac{W_x}{N_{e,x}}N_{e,0} .$$

It follows a slight difference between W_0 and W_m which is determined by

$$\frac{W_0}{W_m}=\frac{1+1.75q_0\alpha_0}{1+1.75q_x\alpha_x} .$$

Theoretical predictions are given in Table III.

TABLE II. Experimental Xe I full half-widths normalized for $N_{e,0}=10^{17}$ cm^{-3} and $T_0=10000$ K from n plasma conditions near T_0.

Transitions	λ(Å)	Richou n	Richou γ_{m1}(Å)	Lesage and Miller n	Lesage and Miller γ_{m2}(Å)	Truong-Bach n	Truong-Bach γ_{m3}(Å)
$6s(\frac{3}{2})2$-$6p'(\frac{1}{2})1$	4500	7	1.4±25%	6	1.28±15%	1	1.34±10%
$-6p'(\frac{3}{2})2$	4524	7	1.1±25%			1	0.82±10%
$-7p\ (\frac{3}{2})2$	4624	9	6.8±15%	4	6.46±15%	1	6.94±10%
$-7p\ (\frac{5}{2})3$	4671	9	5.7±15%	5	5.34±15%	1	5.57±10%
$-7p\ (\frac{5}{2})2$	4697	5	5.5±15%				
$6s(\frac{1}{2})1$-$6p'(\frac{3}{2})2$	4734	6	1.2±25%	4	1.01±15%	1	1.21±10%
$-7p\ (\frac{1}{2})0$	4807					1	3.78±10%
$-7p\ (\frac{3}{2})2$	4843	7	6.9±25%				

These include up to 36 interacting levels.[25] Differences between total Stark and electron-impact widths do not exceed 22% for lines with $\alpha \leq 0.289$. However, for lines corresponding to high ion-broadening asymmetry parameter ($\lambda=4624$ and 4834 Å), ion and electron-impact contributions are of comparable importance. Significant asymmetry of line profiles is observed experimentally for lines corresponding to $\alpha \geq 0.289$.[24] Use of the Griem's approximate formula yields similar results to calculations of the whole profiles found by the Hooper's ion-microfield distribution.

Recommended broadening parameters obtained from weighted averages of three data sets appear in Table IV. These are compared with the results of Klein and Meiners[3] and with theoretical predictions (of the direct calculation method). Ratios of our measured widths to these earlier authors lie between 0.80 and 0.92. This level of agreement is within mutually estimated error. The ratio average of 0.843 is significant to 99% confidence. Also, scatter between our three independent data sets (Table II) is considerably smaller than the scatter between the mean of present values and Klein and

TABLE III. Theoretical Stark parameters for Xe I calculated according to various theories: γ_e, electron-impact full half-widths; α, ion-broadening parameters; γ_0, full total Stark widths. Plasma conditions: $N_{e,0}=10^{17}$ cm^{-3} and $T_0=10000$ K.

Transitions	λ(Å)	Electron-impact γ_e(Å)	α	Total Stark[a] γ_0(Å)
$6s(\frac{3}{2})2$-$6p'(\frac{1}{2})1$	4500	1.310	0.077	1.403
$-6p'(\frac{3}{2})2$	4524	0.902	0.028	0.926
$-7p\ (\frac{3}{2})2$	4624	3.650	0.848	6.556
$-7p\ (\frac{5}{2})3$	4671	3.230	0.289	4.106
$-7p\ (\frac{5}{2})2$	4697	2.660	0.247	3.276
$6s(\frac{1}{2})1$-$6p'(\frac{3}{2})2$	4734	0.976	0.027	1.000
$-7p\ (\frac{1}{2})0$	4807	1.770	0.153	2.024
$-7p\ (\frac{3}{2})2$	4843	4.000	0.848	7.187

[a]Calculated with Griem's formula.

TABLE IV. Comparison between present experimental results (with probable errors for 99%-confident limit), other experiment, and total Stark-width calculations for Xe I full line widths. Plasma conditions: $N_{e,0}=10^{17}$ cm^{-3} and $T_0=10000$ K. s, systematic errors. Results in angstrom units.

Transitions	λ(Å)	Mean of present results	Klein and Meiners (3)	Total[a] Stark
$6s(\frac{3}{2})2$-$6p'(\frac{1}{2})1$	4500	1.34± 4%	1.46±(5.5+s)%	1.41
$-6p'(\frac{3}{2})2$	4524	0.96±24%		0.93
$-7p\ (\frac{3}{2})2$	4624	6.73± 3%	8.20±(4.9+s)%	6.95
$-7p\ (\frac{5}{2})3$	4671	5.53± 3%	6.68±(4.5+s)%	4.16
$-7p\ (\frac{5}{2})2$	4697	5.5 ± 8%		3.31
$6s(\frac{1}{2})1$-$6p'(\frac{3}{2})2$	4734	1.14± 9%	1.42±(4.2+s)%	1.0
$-7p\ (\frac{1}{2})0$	4807	3.78		2.0
$-7p\ (\frac{3}{2})2$	4843	6.9 ±15%		7.6

[a]Calculated with Hooper's ion-microfield distributions.

Meiners's results (Table IV). This may be due in part to the narrowness of Klein and Meiners's spectral range (≈6.4 Å) in relation to xenon Stark widths (up to 8.2 Å, their values), and perhaps also to their fitting to symmetrical Lorentzian shapes. Satisfactory agreement between total Stark-width predictions and experiments is found for all lines except λ=4807 and 4697 Å. It is plausible that redetermination of oscillator strengths involving interacting levels would improve the agreement between theory and experiment for these two lines.

Experimental and calculated total Stark shifts are shown in Table V. For narrow lines (λ=4500, 4524, and 4734 Å) shifts were measured with an optoelectronic comparator. Shifts of broader lines were read from densitometer tracings. Some discrepancies between our values and Klein and Meiners's are evident. The source of disagreement may be that in the earlier work line peaks were deduced from fiber optic channels with spacing of 0.4 Å in first order.[3] Marked differences between theoretical and experimental shifts exist for many Xe I lines. It is suspected that this is due to the ambiguity arising from taking averages of

TABLE V. Experimental and theoretical Xe I Stark shifts normalized for $N_{e,0}=10^{17}$ cm^{-3} and $T_0=10000$ K. Results in angstrom units.

Transitions	λ(Å)	Truong-Bach	Klein and Meiners (3)	Electron impact	Total Stark
$6s(\frac{3}{2})2$-$6p'(\frac{1}{2})1$	4500	0.18±0.02	0.343	0.518	0.599
$-6p'(\frac{3}{2})2$	4524	0.23±0.02		0.195	0.219
$-7p\ (\frac{3}{2})2$	4624	1.4 ±0.3	2.14	0.218	1.715
$-7p\ (\frac{5}{2})3$	4671	1.9 ±0.3	2.43	0.080	0.668
$6s(\frac{1}{2})1$-$6p'(\frac{3}{2})2$	4734	0.14±0.02	0.28	0.180	0.205
$-7p\ (\frac{1}{2})0$	4807	0.9 ±0.3		0.073	0.268

strongly oscillating phases in the S matrix for close encounters.[11]

In conclusion, our present study shows that existing line-broadening theories[8–10] generally agree with experiment for Xe I, a heavy atom; this finding having been extensively tested[1,2] for light elements (H to Cs) with simple atomic structures.

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