Vol. 86 (1994) *ACTA PHYSICA POLONICA A* No. 3

STARK WIDTH MEASUREMENTS OF NEUTRAL ARGON LINES AND COMPARISON WITH ANALOGOUS TRANSITIONS FOR HOMOLOGOUS ATOMS

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(Received March 11, 1994; in final form May 17, 1994)

Half widths of 24 ArI lines from a wide interval of the spectrum and from a large range of excitation energies were measured. A wall-stabilized arc at atmospheric pressure was applied as a plasma generator. Electron densities of the plasma of the order of 10^{16}–10^{17} cm^{-3} were obtained on the basis of hydrogen H_β line broadening measurements. The measured widths of ArI lines, normalized to an electron density of 10^{16} cm^{-3} were compared with experimental data for analogous transitions in other noble atoms.

PACS numbers: 32.70.−n, 32.70.Jz, 52.25.Rv

1. Introduction

Experimental contributions toward understanding of the Stark broadening in noble gases have largely focused on helium, neon and the most economical inert gas — argon. The knowledge of the Stark broadening parameters is essential e.g. for diagnostic purposes, because from the Stark width of a spectral line the electron density of the plasma can be easily deduced. The purpose of this paper is (i) to investigate the Stark broadening of ArI lines (from the visible to near infrared) and consequently from a wide interval of excitation energies (13.3–15.3 eV) and (ii) to compare the measured widths for argon lines with those for analogous transitions in other homologous emitters as neon, krypton and xenon.

2. Experimental

A wall-stabilized arc of the Maecker–Shumaker type [1, 2] at atmospheric pressure was used for generation of the plasma. By changing the arc current and selecting the gas composition ($Ar + H_2$, $Ar + He + H_2$, $Ar + N_2 + H_2$), plasmas of electron densities near the arc axis in the range of 1.4×10^{16} to 1.2×10^{17} cm^{-3} and of temperatures between 9800 and 13400 K, were produced (total number of experiments — 9). The radiation emitted in end-on direction, from the homogeneous plasma volume around the arc axis, was analyzed applying a PGS2 spectrometer, equipped with a photomultiplier. In a few cases a photographical detection of the spectrum was applied.

In the case of photoelectrical detection, the spectral line shapes were measured directly by turning the grating and measuring the photomultiplier output. In a few cases of photographical detection, the measured light transmissions of the photoplate were converted into the intensity scale via the characteristic curve of the photoplate for the given wavelengths.

A low current carbon arc after Euler [3, 4] and/or a calibrated tungsten strip lamp were used as radiation standards. For the purpose of line broadening measurements the knowledge of absolute line intensities was not critical at all. However, the measured intensities of the studied lines in their centres were applied for controlling the possible presence of self-absorption of radiation in the homogeneous plasma layer. For all lines under study, the intensity in the line core was far below the blackbody limit for the corresponding wavelength and plasma temperature, indicating optically thin conditions. The electron densities were obtained from the broadening of the H_β line, applying the data of Kepple and Griem [5, 6]. In the electron density range of this paper, a simplified formula for N_e determination can be used [7]:

$$N_e = 1.01 \times 10^{16} \left(\Delta\lambda_{1/2}\right)^{1.45}, \tag{1}$$

where N_e is obtained in cm^{-3} and $\Delta\lambda_{1/2}$ is the full width at half maximum (FWHM) of the H_β line (in nm).

The temperatures of the plasma were determined on the basis of measured total line intensities (in absolute units) of spectral transitions of neutral elements contained in the plasma (e.g. ArI, NI and HI or ArI and HI) and assuming that the plasma is in local thermal equilibrium at a total pressure of 1 atm. In the case of the Ar + He + H plasma and for other "high temperature" experiments ($T > 12000$ K) the Boltzmann plot method, based on ArII line intensity measurements, was applied.

The accurate knowledge of the plasma temperature was not essential at all for line broadening studies of ArI lines performed in this work. However, the temperature values were used to estimate the small contributions from the Doppler broadening to the measured line widths. The pure Stark widths were evaluated after an appropriate "subtraction" of the Doppler as well as the apparatus broadening, from measured line widths.

3. Results and data reduction

In Table I the Stark widths (FWHM) for 24 ArI lines from the wavelength interval 400–880 nm, determined at different plasma conditions (N_e, T) are listed. The uncertainties of the evaluated Stark widths are mainly around 10%. Only in the case of the lines 629.69, 653.81, 660.48, 826.45 and 852.14 the error may reach the level of 20%. According to the theory [6], the FWHM of a line caused by the

TABLE I

Full (Stark) widths at half maximum for ArI lines (in pm) obtained at various plasma conditions.

Wave-length [nm]	Plasma parameters: N_e in 10^{16} cm^{-3}, T in K								
	12 13200	9.9 12600	8.5 12500	5.1 11500	4.5 13400	4.2 11300	3.1 10800	2.9 10750	1.4 9800
404.44				108	95				30
415.86		200		105	107		60		
416.42				96					30
418.19		245		118	119		75		35
419.10		190		103	117		71		
420.07					94				
425.12							42		
425.94		200		114	108		70		31
426.63		240		105			60		33
427.22		180		100	91		50		28
430.01		180		105	90		62		
433.36					110				
451.07					118		73		
629.69								730	
630.77								510	
653.81								300	
660.48								300	
710.75								320	
712.58								380	
714.70	70								
720.70			1470			660		400	
826.45						38			
852.14						31			
876.17			1180			600			

Stark broadening at an electron density of 10^{16} cm^{-3} can be calculated from the formula

$$\mathrm{FWHM} = 2w_\lambda\,[1 + 1.75\alpha(1 - Cr)]\,, \qquad (2)$$

where $C = 0.75$ for an atomic line and $C = 1.2$ for an ionic one, r is the so-called screening parameter, depending on electron density and temperature of the plasma, w_λ is the electronic broadening parameter, depending mainly on the distance of the levels under consideration to the neighbouring (perturbing) levels, α is the so-called ionic broadening parameter, contributing to the line width and causing an asymmetry of the line profile. The parameter w_λ is proportional to the electron density (N_e), while the parameter α is proportional to $N_e^{0.25}$.

In order to compare the results for a given line at different plasma conditions, and to compare our results with literature data, the measured line widths were normalized to an electron density standard value of $N_e = 10^{16}$ cm^{-3}.Within the normalization procedure the temperature dependencies of $w_\lambda(T)$ and $\alpha(T)$ were neglected. Also the weak dependence of α on electron density ($N_e^{0.25}$) and the very

TABLE II

Comparison of normalized (to $N_e = 10^{16}$ cm^{-3}) Stark widths of Ar I lines obtained in this work with experimental data taken from the literature and with calculated on the basis of theoretical broadening parameters taken from Griem [6].

Wave-length [nm]	Full widths at half maximum (in pm), normalized to $N_e = 10^{16}$ cm^{-3}							
	[8] [9]*	[10] [11]*	[12] [13]*	[14] [15]*	[16] [17]*	[18] [19]*	[6]	This work
404.44		15–16*	16–18	18–22		19		21
415.86	24	19	16–18	18–21	20*			21
416.42			16–18	22		23–25		20.5
418.19		23–25*	19–21	36	23	23–25		25
419.10						19		22
420.07	22		16–17			22	22.5	21
425.12	15		15–17					14
425.94	26	23	19–21	22–29	24		29.4	22
426.63	24		17–20					23
472.22	22	19	16–18	18–24		22	22.4	19
430.01	22	19	16–18	17–22		13–23*		21
433.36	24		17–18			22		24
451.07		25.5	21–23	32–35		27	40.1	24
629.69			335-360				246	256
630.77			180–210					180
653.81			87–91					105
660.48			87–104					105
710.75								112
712.58								133
714.70	6.0*		7.8*	8.0*				5.8
720.70	103*						147	157
826.45	8.6*		9.4*					9.0
852.14			9.5*					7.4
876.15							141	140

weak dependence of the screening parameter on $N_e^{1/6}$ and $T^{-1/2}$ was neglected. In the range of the plasma parameters of this work, all those factors may influence the normalized widths by few percent only. In Table II these normalized FWHM are compared with the data taken from the literature and calculated according to Griem [6].

TABLE III

Full widths at half maximum of the studied ArI lines in frequency units (rad/s) normalized to an electron density of 10^{16} cm^{-3}.

Wave-length [nm]	Transition	J_m–J_n	Excitation energies ($E_m - E_n$) [eV]	$(\chi - E_n)^{-1}$ [eV^{-1}]	$\Delta\omega_{1/2}$ [10^{11} rad/s]
714.70	$4s[3/2]$–$4p'[3/2]$	2–1	11.55–13.28	0.377	0.21
852.14	$4s'[1/2]$–$4p'[3/2]$	1–1	11.83–13.28	0.377	0.22
826.45	$4s'[1/2]$–$4p'[1/2]$	1–1	11.83–13.33	0.384	0.29
425.12	$4s[3/2]$–$5p[1/2]$	2–1	11.55–14.46	0.772	1.5
420.07	$4s[3/2]$–$5p[5/2]$	2–3	11.55–14.50	0.797	2.2
430.01	$4s[3/2]$–$5p[5/2]$	1–2	11.62–14.51	0.803	2.0
416.42	$4s[3/2]$–$5p[3/2]$	2–1	11.55–14.52	0.810	2.2
427.22	$4s[3/2]$–$5p[3/2]$	1–1	11.62–14.52	0.810	1.95
415.86	$4s[3/2]$–$5p[3/2]$	2–2	11.55–14.53	0.816	2.3
426.63	$4s[3/2]$–$5p[3/2]$	1–2	11.62–14.53	0.816	2.4
451.07	$4s'[1/2]$–$5p[1/2]$	1–0	11.83–14.58	0.851	2.2
419.10	$4s'[1/2]$–$5p'[3/2]$	0–1	11.72–14.66	0.786	2.35
418.19	$4s'[1/2]$–$5p'[1/2]$	0–1	11.72–14.69	0.805	2.7
404.44	$4s[3/2]$–$5p'[3/2]$	1–2	11.62–14.69	0.805	2.5
433.36	$4s'[1/2]$–$5p'[3/2]$	1–2	11.83–14.69	0.805	2.3
425.94	$4s'[1/2]$–$5p'[1/2]$	1–0	11.83–14.74	0.839	2.3
876.15	$4p'[1/2]$–$4d[3/2]$	1–2	13.33–14.74	0.985	3.4
710.75	$4p[5/2]$–$6s[3/2]$	2–2	13.09–14.84	1.09	4.2
653.81	$4p[5/2]$–$4d'[3/2]$	3–2	13.08–14.95	1.02	4.6
660.48	$4p[5/2]$–$4d'[3/2]$	2–3	13.09–14.97	1.04	4.5
712.58	$4p'[3/2]$–$6s'[1/2]$	1–1	13.28–15.02	1.10	5.0
720.70	$4p'[3/2]$–$6s'[1/2]$	2–1	13.30–15.02	1.10	5.7
630.77	$4p[3/2]$–$5d[3/2]$	2–2	13.17–15.14	1.63	8.5
629.69	$4p'[1/2]$–$5d'[3/2]$	1–2	13.33–15.30	1.58	12.0

In Table III the studied lines are listed in an order, determined by the excitation energy of the upper level (E_n) of the corresponding transition. In the 5th column the quantity $(\chi - E_n)^{-1}$ in eV^{-1} is listed, where χ is the energy limit of the specific ArI level system: 15.76 eV and 15.94 eV for the two systems under consideration, respectively. In the last column the corresponding line widths are listed in ω units (rad/s), evaluated according to the formula

$$\Delta\omega_{1/2} = 2\pi c\lambda^{-2}\Delta\lambda_{1/2}. \tag{3}$$

In Fig. 1 these widths (in frequency units) are plotted as a function of the reciprocal binding energy $(\chi - E_n)^{-1}$. The full circles and solid line represent the

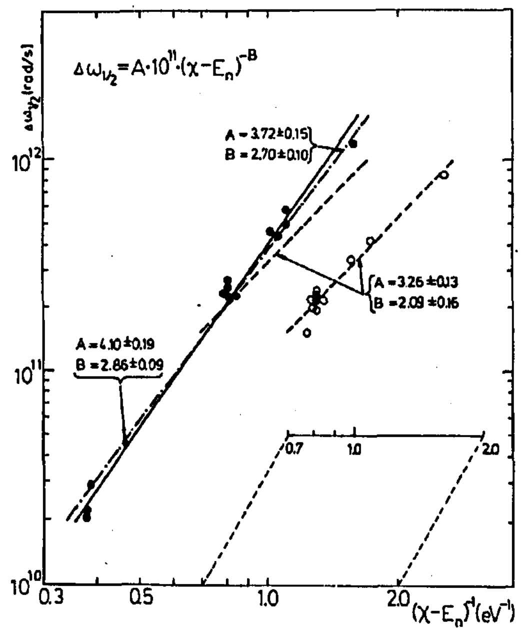

Fig. 1. The experimental Stark FWHM (in frequency units) of ArI lines, normalized to an electron density value of 10^{16} cm^{-3}, versus the reciprocal binding energy of the upper levels from which the emission lines originate. The full circles and the solid line correspond to spectral lines arising from primed upper levels, while the open circles and the dashed line correspond to the spectral lines arising from non-primed upper levels. The dotted-dashed line represents the best fit, when results for all studied lines are approximated by a formula of the type $\Delta\omega_{1/2} = A(\chi - E_n)^{-B}$.

results for lines originating from primed upper levels, while the open circles and the dashed line represent the results for lines with non-primed upper levels. The results of both line groups (level systems) are well fitted by formulas of the type, proposed by Purić et al. [20]:

$$\Delta\omega_{1/2} = A(\chi - E_n)^{-B}. \tag{4}$$

The corresponding values for A and B are quoted in the figure. If both groups of lines (both level systems) are taken together, a best fit for $A = (3.72 \pm 0.15) \times 10^{11}$ and $B = 2.70 \pm 0.10$ is obtained (dashed-dotted line).

As one can see, the formulas can be used with confidence for estimation of the widths for ArI spectral lines with unknown broadening parameters.

4. Comparison of results obtained for argon with literature data for homologous atoms

The experimental data of this work support the well-known fact that in most cases the main contribution to the Stark width of a spectral line arises from the broadening of the upper level of the respective transition. Indeed, the normalized Stark widths determined in this work scale, as expected, with the upper level ionization energy according to Eq. (4). The width of the upper level itself is determined by its "interaction" with neighbouring perturbing levels [6]. Because of similarities of the energy level system of inert gases, one can also expect some regular behaviour of the Stark broadening parameters for analogous transition, along the atomic number of noble emitters. Recently, such regularities have been reported e.g. by Di Rocco [31] and Bertuccelli and Di Rocco [32] for singly ionized inert gases and Djeniže et al. for singly ionized elements of the second group in the periodic system [33].

Therefore, our results for argon are compared with data available in the literature for Ne, Kr and Xe. Unfortunately, for only 13 of the studied Ar lines, data for analogous transitions in other noble gases could be found: 6 for NeI (Refs. [21–23] and [29]), 3 for KrI (Refs. [24, 27] and [28]), and 7 for XeI (Refs. [24–26] and [30]).

In Table IV the FWHM (in frequency units), normalized to a common electron density value of 10^{16} cm^{-3}, originating from homologous upper levels of Ne, Ar, Kr and Xe and the corresponding ionization energies are listed. In Table V similar data for the remaining analogous transitions in Ar, Kr and Xe are presented. As one can see, for almost all these transitions a clear tendency could be revealed: the normalized line width (in frequency units) increases with decreasing ionization energy of the upper level of the transition under consideration. Only the results obtained by Purić et al. [21] for NeI do not follow this rule. These line broadening data are systematically too large, not only compared with our results for ArI, but also compared with other experimental data obtained by Nubbemeyer et al. [23] and Döhrn and Helbig [29].

Unfortunately, only for the transition array ns–np' ($n = 3$ for Ne, $n = 4$ for Ar etc.) results for all 4 elements (Ne, Ar, Kr and Xe) are available. For neon and argon, additional results for the transition ns'–np' are reported. These results for 4 NeI lines, 3 ArI lines and one for KrI and XeI are compared in Table IV. All these lines originate from common upper levels $np'[1/2]_1$ or $np'[3/2]_1$. As can be seen, with increasing atomic number (Z) the bond energy (or ionization energy) of the upper level np' decreases systematically from 2.97 eV for Ne to 2.39 eV for Xe.

In Fig. 2 two quantities, as a function of the inverse ionization energy of the upper level, are plotted:

1. below the solid border line connecting the element symbols — the normalized FWHM $\Delta\omega_{1/2}$ in frequency units, and
2. above the solid line — the same widths ($\Delta\omega_{1/2}$) multiplied by $Z^{1/2}$, where Z is the atomic number of the emitter.

The second quantity is a particular form of a more general expression $Z^{l>/2}$, which has been found to fit very well the data obtained for singly ionized noble atoms [32]. The symbol $l>$ in the exponent is the greater orbital quantum number of the studied transition — in our case for the transition $s(s')$–p', $l> = 1$.

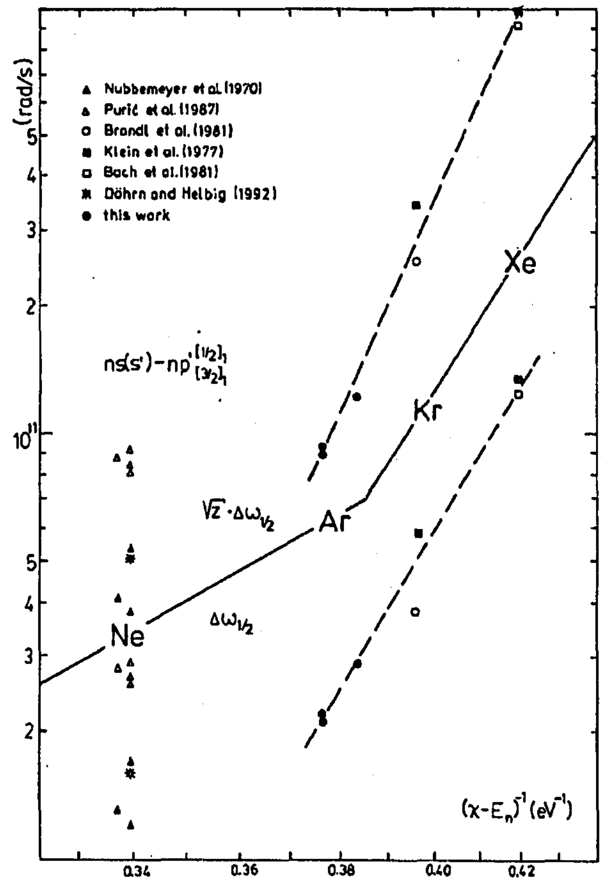

Fig. 2. Comparison of broadening data for spectral lines of noble gases. Two quantities as a function of the inverse ionization energy of the upper level are shown: (1) below the solid line connecting the element symbols — the normalized FWHM $\Delta\omega_{1/2}$, and (2) above the solid line — the same widths multiplied by the square root of the atomic number of the emitter.

TABLE IV

Full widths at half maximum (in frequency units) normalized to an electron density of $N_e = 10^{16}$ cm^{-3} for spectral lines of Ar, Kr and Xe, originating from homologous upper levels, characterized by their individual ionization energies I_n in eV.

Element	Binding energy I_n/eV	Designation of the upper level $4p'[3/2]_1$	$4p'[1/2]_1$	$4d[3/2]_2$	$6s'[1/2]_1$
Ne	2.97	0.28[a]			
		0.13[c]			
	2.94		0.26[a]		
			0.27[a]		
			0.29[a]		
			0.17[c]		
			0.12[c]		
			0.16[i]		
	1.52			1.4[a]	
	1.00				3.3[b]
Ar	2.65	0.21			
		0.22			
	2.60		0.29		
	1.02			3.4	
	0.91				5.7
					5.0
Kr	2.52		0.58[d]		
			0.39[g]		
Xe	2.39		1.36[d]		
			1.25[e]		

[a] Ref. [21] [b] Ref. [22] [c] Ref. [23] [d] Ref. [24] [e] Ref. [25] [g] Ref. [27]
[i] Ref. [29]

As can be seen, the results for Ar, Kr and Xe are well fitted by the formula $\Delta\omega_{1/2} = A_1 I^{-B_1}$ as well as by the relation $Z^{1/2}\Delta\omega_{1/2} = A_2 I^{-B_2}$.

Only the widths reported for NeI lines do not fit the "regularity trends". The results for Ne I obtained by Purić et al. [21] are approximately 2 times larger than the "old" data of Nubbemeyer et al. [23]. These "old" data seems to be much more reliable and have been recently confirmed in the experiment of Döhrn and Helbig [29]. Even ignoring the results of Purić et al., the deviation of the Ne data from the regularity trends for homologous atoms is very large.

TABLE V

Full widths at half maximum (in frequency units) normalized to an electron density of $N_e = 10^{16}$ cm^{-3} for spectral lines of Ar, Kr and Xe, originating from homologous upper levels, characterized by their individual ionization energies I_n in eV.

Element	Binding energy I_n/eV	Designation of the upper level				
		$5p[1/2]_0$	$5p[3/2]_2$	$5p[3/2]_1$	$5p[5/2]_2$	$5p[5/2]_3$
Ar	1.18	2.2				
	1.22		2.3			
		2.4				
	1.23			2.2		
				1.95		
	1.24				2.0	
	1.25					2.2
Kr	1.14	4.1[d]				
	1.22		3.0[d]			
				2.7[h]		
Xe	1.12	3.1[e]				
	1.13			3.85[d]		
	1.14		5.9[e]			
			5.5[e]			
			7.2[d]			
			6.7[j]			
	1.16					4.8[e]
						5.8[d]
						5.6[j]
	1.18			4.8[f]		
				4.7[e]		

[d] Ref. [24] [e] Ref. [25] [f] Ref. [26] [h] Ref. [28] [j] Ref. [30]

This departure of results for neon from the regularity rule can be explained on the basis of detailed analysis of regularities of the energy level system in noble gases [34]. Indeed, the energy levels responsible for the emission of the studied lines and the levels which "interact" with the levels of interest of Ar, Kr and Xe show clear regular trends, while the corresponding Ne levels reveal distinct dissimilarities.

5. Conclusions

Line broadening parameters (the Stark widths) for an extensive set of ArI lines determined in this work verify the characteristical dependencies of the Stark widths on the upper level ionization energy. This regularity observed in ArI spectrum can be used for estimation of the Stark broadening parameters of lines, for which no data are reported in the literature. Comparison of results obtained for ArI with data for analogous transitions in other noble gases, reveals typical regularity of the Stark width dependence on the upper level ionization energy in the sequence of atoms Ar–Kr–Xe. Deviations from the regular trend for Ne lines were found, which can be attributed to some peculiarities of the energy level system of neon.

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