

### Stark broadening and regularities of prominent multiply ionized nitrogen spectral lines

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Stark widths of four N II, fourteen N III, five N IV, and two N V spectral lines have been measured in a linear-pinch discharge plasma and compared with existing experimental and theoretical results. Electron densities determined with single-wavelength laser interferometry with use of the visible 632.8-nm transition of a He-Ne laser cover the range of  $(0.40-1.80) \times 10^{23} \text{ m}^{-3}$ . The electron temperature of 50 000 K was determined from the Boltzmann slope of several N III spectral lines, and from the relative intensity ratios of several N II to N III and N III to N IV spectral lines. Since the majority of investigated N III spectral lines originate from the  $3s-3p$  and  $3p-3d$  transition arrays, the Stark-width ( $w$ ) dependence on the upper level ionization potential ( $I$ ) of corresponding lines belonging to these two transition arrays was discussed and found to be of the form  $w = aI^{-b}$ , where  $a$  and  $b$  are constants independent of the ionization potential. Similarly, the overall Stark width trends, using data obtained from the  $3s-3p$  transition of the N II, N III, N IV, and N V spectral lines, were determined and discussed including the influence of emitter net charge on the Stark broadening dependence on the upper-level ionization potential. The established overall trend was used to predict the Stark width of uninvestigated spectral lines originating from the given  $3s-3p$  transition with an accuracy better than 30%.

#### I. INTRODUCTION

A number of papers<sup>1-10</sup> deals with Stark-width measurements of N II spectral lines originating from different multiplets. However, only two papers<sup>1,2</sup> were devoted to the Stark-shift measurements. The obtained Stark-width and -shift data were compared with the experiment/theory ratios using the tabulation given by Griem and co-workers<sup>11,12</sup> within semiclassical approximation and by Dimitrijević and Kojić<sup>13,15</sup> within a modified

Griem's<sup>14</sup> semiempirical approach (see also Refs. 16 and 17). In general, the results of these experiments agree well with each other (within an accuracy of  $\pm 20\%$ ), with several exceptions due to the inhomogeneities,<sup>1</sup> self-absorption in the line profile measurements,<sup>9</sup> or other sources problems.<sup>18</sup>

Several experimental<sup>6,8-10</sup> and theoretical<sup>13-15</sup> papers were devoted to the investigation of the Stark broadening of the most prominent doubly ionized nitrogen spectral lines. Large discrepancies were found to exist among the experimental results of Popović *et al.*,<sup>6</sup> Källne *et al.*,<sup>8,9</sup> and Purcell and Barnard.<sup>10</sup> All calcula-

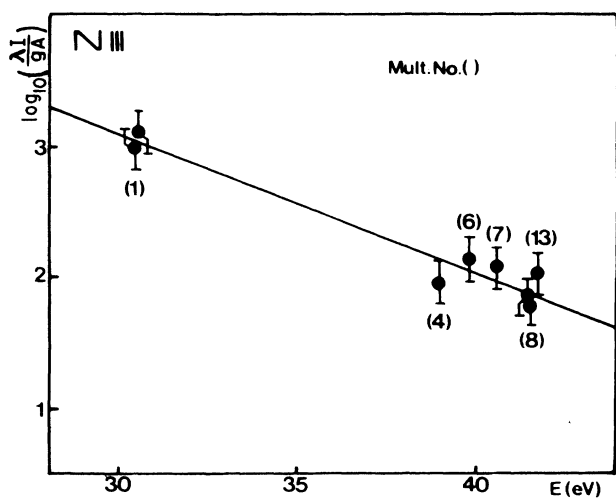


FIG. 1. Plot of  $\log(I\lambda/gA)$  against excitation energies of upper levels for N III lines. The intensity  $I$  is measured in arbitrary units at  $2.5 \mu\text{s}$  after the beginning of the discharge.

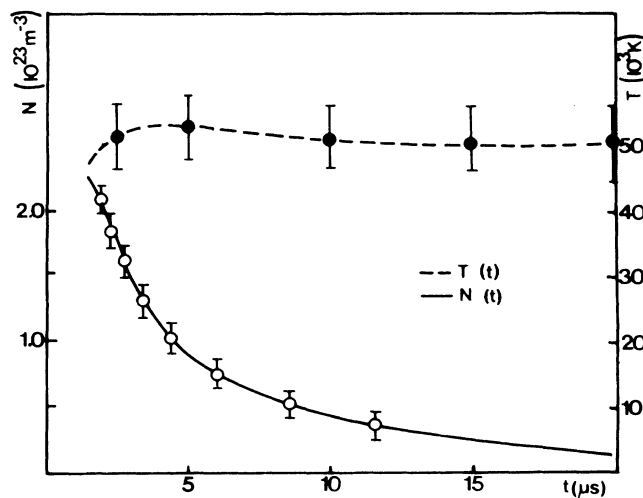


FIG. 2. Observed electron density and temperature decays.

tions were done in the framework of Griem's semiempirical approach<sup>19</sup> with several modifications<sup>13-15</sup> undertaken in order to improve the agreement with the experimentally obtained results (within  $\pm 10\%$ ). The only experimental result available for the N IV 405.77-nm spectral line is given by Källne *et al.*<sup>8</sup> and was found to be 5.00 times larger than the result of the modified semiempirical approximation obtained by Dimitrijević and Konjević.<sup>13-15</sup> There are no published results (either experimental or theoretical) for any NV spectral lines known to the authors. Therefore, the aim of this paper was to supply more experimental Stark broadening data for N II, N III, N IV, and NV spectral lines and to compare them with the existing experimental and theoretical data.

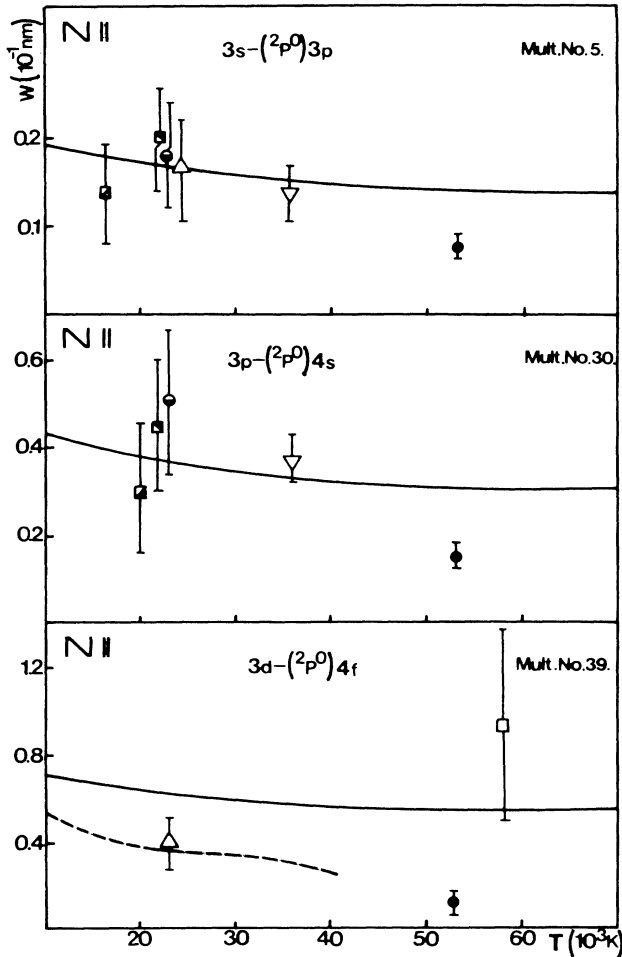


FIG. 3. Stark HWHM's of N II spectral lines vs electron temperatures:  $\blacksquare$ , Day and Griem (Ref. 1);  $\blacksquare$ , Berg *et al.* (Ref. 2);  $\bullet$ , Jalufka and Craig (Ref. 3);  $\square$ , Konjević *et al.* (Ref. 4);  $\triangle$ , Popović *et al.* (Ref. 6);  $\square$ , Källne *et al.* (Ref. 8);  $\nabla$ , Purcell and Barnard (Ref. 10);  $\bullet$ , our experimental data; —, Jones *et al.* (Ref. 12) semiclassical theoretical data; and - - -, Dimitrijević and Konjević (Ref. 14) modified semiempirical results.

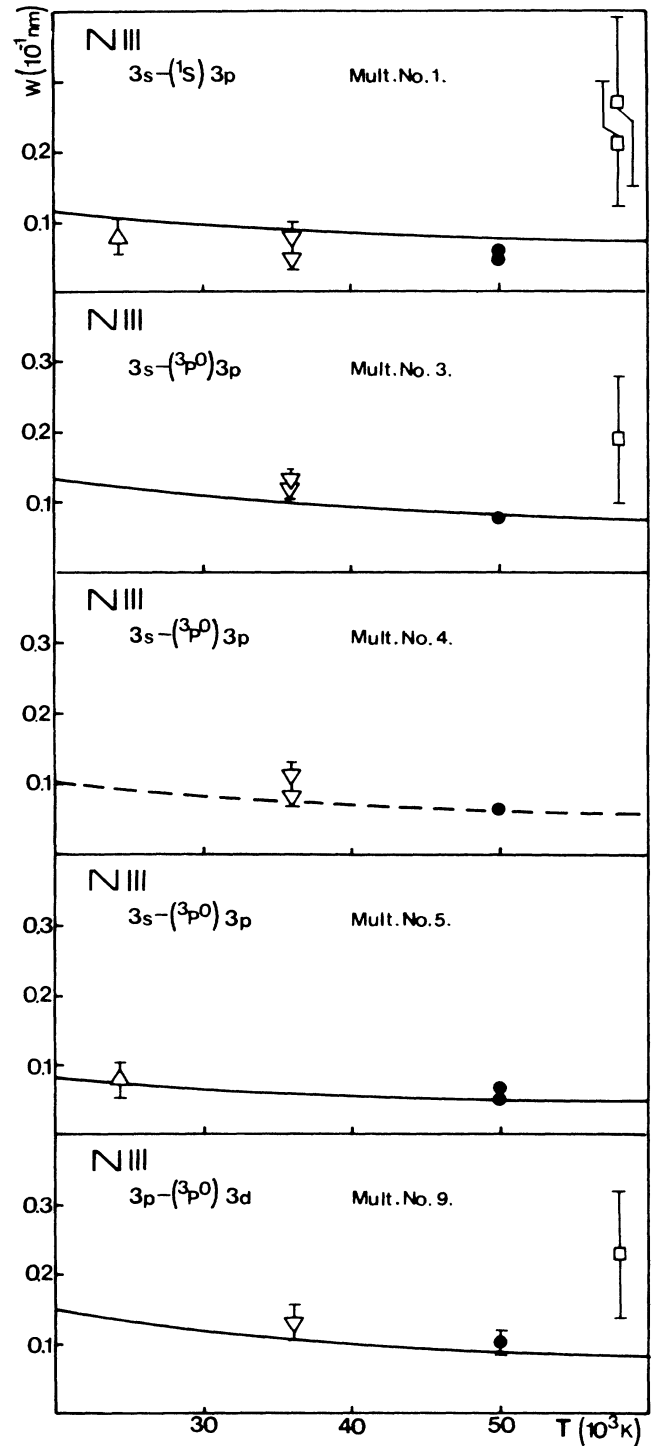


FIG. 4. Stark HWHM's of N III spectral lines vs electron temperatures:  $\triangle$ , Popović *et al.* (Ref. 6);  $\square$ , Källne *et al.* (Ref. 8);  $\nabla$ , Purcell and Barnard (Ref. 10); and  $\bullet$ , our experimental data. —, modified semiclassical theoretical results (Ref. 14), and - - -, predicted values obtained using Eq. (1) corrected to the temperature dependence.

Recently, interest has grown for better understanding of Stark broadening regularities within an analyzed spectrum.<sup>20,21</sup> The majority of investigated N II, N III, N IV, and N V spectral lines originates from the  $3s-3p$  and  $3p-3d$  transition arrays. Therefore, it was possible to discuss the feature of Stark broadening dependence on the upper-level ionization potential. The obtained dependence within the N III spectrum is of the same kind as that previously obtained<sup>20,21</sup> in case of Ne I and Ne II spectral lines originating from  $3s-3p$  and  $3p-3d$  transition arrays. At the same time we investigated the influence of the emitter net charge on the Stark broadening of the line originating from the  $3s-3p$  transition arrays among four different stages of ionization (N II, N III, N IV, and N V). The obtained functional dependences have been used for predicting the Stark-width values for several investigated spectral lines where there are no other calculated results. The overall agreement of these predicted values with the experimentally obtained ones is within  $\pm 30\%$ . This can be regarded as a very good agreement, taking into account that the accuracy of semiempirical or any other theoretical approach in Stark-width calculations in case of highly striped ions is of the same order.

## II. EXPERIMENTAL APPARATUS AND PROCEDURE

All measurements of the line profiles were made in a linear plasma source and the same scanning procedure is used as described elsewhere.<sup>21</sup> Here the differences are only given for the sake of completeness. The working gas was nitrogen at 67 Pa pressure. The discharge current is supplied by a capacitor of 0.3  $\mu\text{F}$  charged to 15 kV giving a current with a maximum up to  $I_m = 8.2$

kA and period  $T \simeq 3 \mu\text{s}$ . For this experiment we used a photomultiplier (EMI 9789 QB) and grating spectrograph (Zeiss PGS-2, inverse linear dispersion in the double pass 0.367 nm/mm) system. The scanning of the lines was done by a shot-to-shot technique advancing the exit slit-photomultiplier combination in small-wavelength steps. Another monochromator (Zeiss SPM-1) with a photomultiplier was simultaneously used to monitor the continuum radiation from the same part of the plasma as a check for shot-to-shot reproducibility. The obtained profiles were of a Voigt type as a result of convolution of the Lorentzian Stark profile and the Gaussian profile due to instrumental and Doppler broadening. Van der Waals and resonance broadening were found to be negligible.

Therefore, an instrumental half width at half maximum (HWHM) of 0.004 nm and Doppler HWHM of corresponding spectral lines were considered as the competing fractions in comparison with Stark broadening in the total line HWHM. The standard deconvolution procedure was used.<sup>22</sup> Great care was paid to minimize the influence of self-absorption on the Stark-width determination. Opacity was checked by measuring line-intensity ratios within the same multiplet. The obtained values were compared with calculated ratios of the products of the spontaneous emission probabilities and the corresponding statistical weights<sup>23</sup> of the corresponding upper level of the investigated spectral lines. These ratios were found to be the same (within  $\pm 10\%$ ) within each multiplet. In this experiment reproducibility of the plasma was simultaneously controlled by measuring the intensity of continuum radiation at a chosen wavelength ( $\lambda$  is less than the blue wing of the Ne I 632.8-nm spectral line) and was found to be within  $\pm 7\%$ .

TABLE I. Stark HWHM's  $w_m$ —our experimental and the results of other authors (Refs. 1–10); ratios of measured to theoretical results ( $w_{\text{th1}}$ ) of Griem and co-workers (Refs. 11 and 12) are given. The electron density and temperature, multiplet number, and wavelengths are also given.  $w_{\text{pr1}}$  are the predicted values according to Eq. (3).

Emitter ion	Transition array	Multiplet (No.)	Wavelength		$T$ ( $10^4$ K)	$N$ ( $10^{-23}$ m $^{-3}$ )	$W_m$ ( $10^{-1}$ nm)	$W_m/W_{\text{th1}}$	$W_{\text{pr1}}$ ( $10^{-1}$ nm)	Ref.
			$\lambda$ (nm)							
N II	$2p\ 3s-$ $2p(^2P^\circ)3p$	$^3P^\circ-^3P$ (5)	463.05	5.30	0.90	0.070		0.057	a	
				3.60	2.90	4.06			10	
				2.20	1.00	0.20	1.18	2		
				2.28	1.00	0.18	1.03	3		
				1.62	3.12	0.44	0.79	4		
				2.315	0.48	0.08	0.99	6		
		$^1P^\circ-^1S$ (13)	343.72	5.30	0.90	0.042		0.035	a	
	$2p\ 3p-$ $2p(^2P^\circ)4s$	$^3P-^3P^\circ$ (30)	383.84	5.30	0.90	0.161	0.44	a		
				1.96	3.30	0.95	0.76	1		
				2.20	1.00	0.44	1.14	2		
			2.28	1.00	0.50	1.32	3			
	$2p\ 3d-$ $2p(^2P^\circ)4f$	$^3P^\circ-^3G$ (39)	404.10	5.30	0.90	0.235	0.48	a		
				2.315	0.48	0.20	0.68	6		

<sup>a</sup>This work.

The electron temperature was determined from: the Boltzmann slope, given in Fig. 1. of eight N III spectral lines (409.73, 410.34, 377.11, 420.00, 334.28, 393.44, 393.85, and 454.64 nm with a corresponding upper-level energy interval of 11.22 eV); the Boltzmann slope of five

N IV spectral lines (347.87, 348.49, 405.78, 346.34, and 374.77 nm with a corresponding upper-level energy interval of 11.63 eV) and from intensity ratios of several N II to N III, and N III to N IV spectral lines, with estimated errors of  $\pm 10\%$ . Transition probability values

TABLE II. Comparison of measured  $w_m$  and calculated Stark HWHM's of N III, N IV, and N V spectral lines. The electron density and temperature, multiplet numbers, and wavelengths are given. Ratios of measured to theoretical results are given in the following orders:  $w_{th2}$  — Dimitrijević and Konjević (Refs. 13–15) modified semiempirical calculation and  $w_{th3}$  calculated values according to Eqs. (11)–(15) from Ref. 13. Predicted values:  $w_{pr1}$  are the predicted values calculated using the Eq. (3); and  $w_{pr2}$  are the predicted values using the Eqs. (1) and (2).

Emitter ion	Transition array	Multiplet (No.)	Wavelength		$T$ ( $10^4$ K)	$N$ ( $10^{-23}$ m $^{-3}$ )	$W_m$ ( $10^{-1}$ nm)	$W_m/W_{th2}$	$W_m/W_{th3}$	$W_{pr1}$ ( $10^{-1}$ ns)	$W_{pr2}$ ( $10^{-1}$ nm)	Ref.		
			$\lambda$ (nm)											
N III	3s- ( $^1P$ )3p	$^2S\text{-}^2P^\circ$ (1)	409.73		5.00	1.78	0.107	0.79	0.73	0.092		a		
					3.60	2.90	0.145	0.57	0.57		10			
					5.80	14	3.8	3.80	3.39		8			
					2.43	0.55	0.048	0.82	0.84		6			
				410.34		5.00	1.78	0.094	0.62	0.64	0.092		a	
						3.60	2.90	0.230	0.90	0.90		10		
						5.80	14	2.9	2.89	2.59		8		
		2s2p3s- 2s2p( $^3P^\circ$ )3p	$^4P^\circ\text{-}^4D$ (3)	451.49		5.00	1.78	0.138	0.92	0.84	0.210	0.146		a
					3.60	2.90	0.375	1.72	1.40		10			
			$^4P^\circ\text{-}^4S$ (4)	377.11		5.00	1.78	0.114		0.156	0.115		a	
						3.60	2.90	0.325					10	
			$^4P^\circ\text{-}^4P$ (5)	336.74		5.00	1.78	0.099	1.03	0.90	0.130	0.101		a
						2.43	0.55	0.049	1.18	1.26			6	
				336.58		5.00	1.78	0.111	1.15	1.01	0.130	0.101		a
			$^2P^\circ\text{-}^2D$ (6)	420.00		5.00	1.78	0.197	1.14	1.04	0.215	0.177		a
						5.80	14	3.7	2.98	2.49			8	
		$^2P^\circ\text{-}^2S$ (7)	334.28		5.00	1.78	0.122			0.151	0.138		a	
	2s2p3p- 2s2p( $^3P^\circ$ )3d	$^2P\text{-}^2D^\circ$ (8)	393.85		5.00	1.78	0.168				0.153		a	
				393.44		5.00	1.78	0.168				0.153		a
		$^2P\text{-}^2P^\circ$ (25 UV)	298.36		5.00	1.78	0.101				0.128		a	
		$^4D\text{-}^4F^\circ$ (9)	486.13		5.00	1.78	0.188	1.16	0.88		0.197		a	
		$^4S\text{-}^4P^\circ$ (13)	454.64		5.00	1.78	0.233				0.219		a	
	4d- ( $^1S$ )5f	$^2D\text{-}^2F^\circ$ (16)	400.36		5.00	1.78	0.512						a	
N IV	2s3s- 2s( $^2S$ )3p	$^3S\text{-}^3P^\circ$ (1)	347.87		5.00	1.78	0.096	1.08	1.28				a	
					5.00	1.78	0.096	1.08	1.28				a	
		2p3s- 2p( $^2P^\circ$ )3p	$^3P^\circ\text{-}^3P$ (7)	346.34		5.00	1.78	0.097			0.130		a	
					$^1P^\circ\text{-}^1D$ (8)	374.77		5.00	1.78	0.131			0.157	
		3s3p- 2s( $^2S$ )3d	$^1P^\circ\text{-}^1D$ (3)	405.78		5.00	1.78	0.131	1.31	0.93				a
						5.80	14	3.7	5.0	4.6			8	
N V	3s-3p	$^2S\text{-}^2P^\circ$ (1)	460.38		5.00	1.78	0.169			0.170			a	
					5.00	1.78	0.169			0.170			a	

<sup>a</sup>This work.

were taken from Wiese *et al.*<sup>23</sup> The electron densities were determined by single-wavelength laser interferometry using the visible transition of a He-Ne laser with 7% precision and was found to be in the range  $(0.40-1.80) \times 10^{23} \text{ m}^{-3}$ . The corresponding electron density decay is given in Fig. 2, together with the electron temperature time dependence. It is found that the temperature is almost constant and equal to  $50\,000 \text{ K} \pm 10\%$  in the covered range of the measured electron density.

### III. EXPERIMENTAL RESULTS AND DISCUSSION

Experimentally determined Stark half widths at half maximum ( $w$ ) of the investigated N II, N III, N IV, and N V spectral lines are given in Tables I and II, together with semiclassical<sup>11,12</sup> and modified semiempirical theoretical calculations,<sup>13-15</sup> experimental data<sup>1-10</sup> from other authors, our theoretically predicted values ( $w_{pr}$ ) using the established trends (dependences on the upper-level ionization potential), and the main plasma parameters, i.e., electron density and electron temperature. Estimated errors of our experimental data given in Tables I and II are as follows: electron density,  $\pm 7\%$ ; electron temperature,  $\pm 10\%$ ; and line HWHM,  $\pm 15\%$ . Results of several N II and N III multiplets are also graphically presented in Figs. 3 and 4 as function of electron temperature for the sake of better comparison with Griem's results,<sup>11,12</sup> modified semiclassical and modified semiempirical calculations,<sup>13-15</sup> and other experimental results.<sup>1-10</sup> From Fig. 3 one can conclude that our experimental Stark-width results for N II spectral lines are smaller than theoretical ones<sup>11,12</sup> for about 50%.

Agreement of our Stark widths of N III spectral lines with data of Popović *et al.*<sup>6</sup> is within  $\pm 10\%$ . However, a large discrepancy exists with the experimental data of Källne *et al.*,<sup>8,9</sup> i.e., for N III spectral lines up to a factor of 3 (which was corrected in paper by Purcell and Barnard<sup>10</sup> to 30% maximum) and for N IV up to a factor of 4. Good agreement has been found with modified semiempirical calculation<sup>13-15</sup> (within  $\pm 20\%$  for N III spectral lines and  $\pm 30\%$  for N IV 405.78-nm spectral line). Unfortunately, there are no experimental or theoretical data for any N V spectral lines and several of the N II, N III, and N IV spectral lines. Therefore, we used the obtained Stark-width dependence on the upper-level ionization potential within investigated  $3s-3p$  and  $3p-3d$  to predict Stark-width values of several N III spectral lines. However, for N IV and N V spectral lines originating from  $3s-3p$  transition arrays the overall trend has been used after including the Stark-width dependence on the net charge of the corresponding emitter (discussed in Sec. IV of this paper). The predicted values ( $w_{pr}$  in Tables I and II) agree with our experimental data within  $\pm 25\%$  for N II,  $\pm 30\%$  for N III,  $\pm 25\%$  for N IV, and  $\pm 2\%$  for N V spectral lines. This can be regarded as very satisfactory since the experimental errors in Stark width are within  $\pm 15\%$ ; and the uncertainty of the discussed theoretical approaches is within  $\pm 30\%$ .

### IV. REGULARITIES AND SYSTEMATIC TRENDS

It has been found that the measured Stark-width data of N III at an electron temperature of  $50\,000 \text{ K}$  and electron density of  $10^{23} \text{ m}^{-3}$ , within the  $3s-3p$  and  $3p-3d$  transition arrays, satisfy the following relations, respectively:

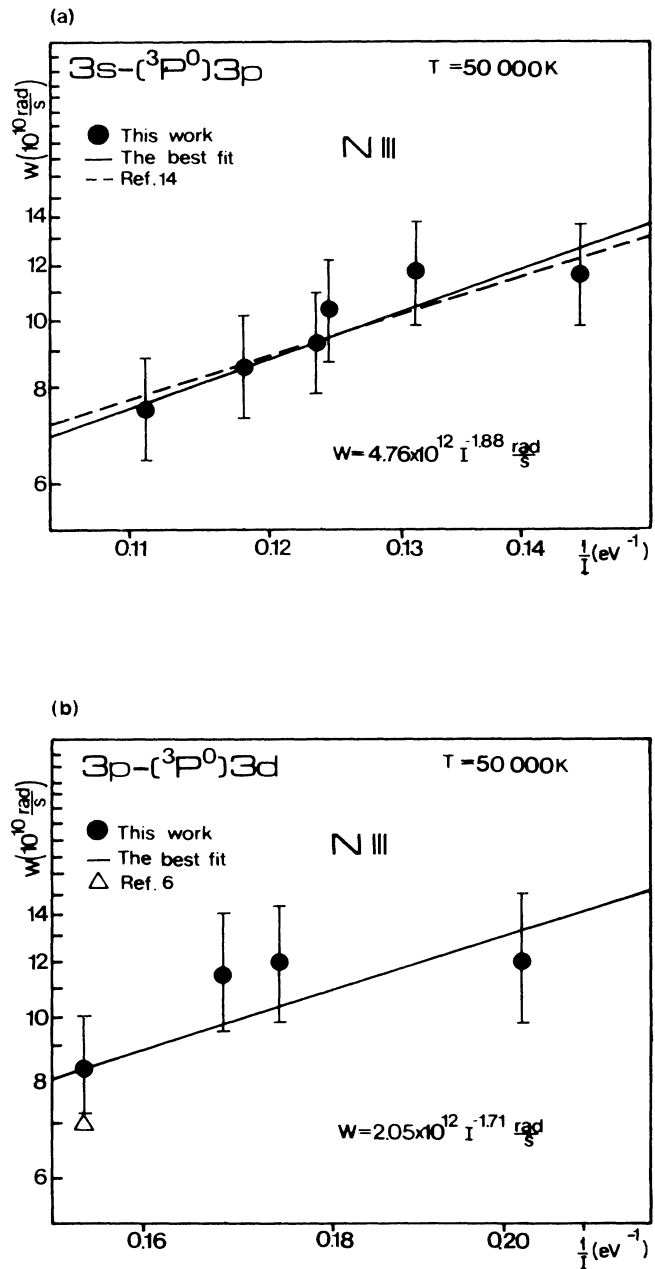


FIG. 5. Stark HWHM ( $w$ ) vs inverse value of the upper-level ionization potential ( $I$ ) for N III (a)  $3s-3p$  and (b)  $3p-3d$  transition arrays at electron temperature  $T = 50\,000 \text{ K}$  and electron density  $N = 10^{23} \text{ m}^{-3}$ .

$$w_{3s-3p} = 4.76 \times 10^{12} I^{-1.88}, \quad (1)$$

$$w_{3p-3d} = 2.05 \times 10^{12} I^{-1.71}, \quad (2)$$

where the ionization potential ( $I$ ) has to be taken in eV to obtain the Stark width ( $w$ ) in angular frequency units. The corresponding correlation factors are 0.80 and 0.65, respectively. Our experimental data and the modified semiempirical theoretical results of Dimitrijević and Konjević<sup>13-15</sup> follow the trend given by Eqs. (1) and (2) within  $\pm 15\%$ . This is shown in Figs. 5(a) and 5(b), which is within an experimental error of  $\pm 15\%$  and a theoretical uncertainty of the semiempirical approach. We have calculated the Stark widths of lines that have not been investigated before, experimentally or theoretically (N III multiplets: 4, 5, 7, 8, 13, and 25 UV) using corresponding relations, Eqs. (1) and (2). The predicted Stark widths ( $\omega_{pr}$ ) thus obtained are found to be in a very good agreement with experimentally observed values (within  $\pm 15\%$ ). There are no other experimental or theoretical results available for the N IV and N V spectral lines from the  $3s-3p$  transition arrays. Therefore, we have constructed the overall trend given in Fig. 6. We have found the following relation for the trend of the Stark-width data for all spectral lines originating from the  $3s-3p$  transition arrays of N II, N III, N IV, and N V:

$$w_{3s-3p} = 7.76 \times 10^{10} Z^2 I^{-0.86}, \quad (3)$$

where  $Z$  is the net emitter charge in atomic units (correlation factor 0.88). We have calculated the Stark-width values of the corresponding N II, N III, N IV, and N V spectral lines using Eq. (3) and found them to be in fairly good agreement with experimental results, i.e., within  $\pm 25\%$  for N II,  $\pm 30\%$  for N III,  $\pm 25\%$  for N IV, and  $\pm 2\%$  for N V. Therefore, Eqs. (1)–(3) can be used for theoretical prediction of Stark-width values for uninvestigated spectral lines but belonging to the corresponding  $3s-3p$  and  $3p-3d$  transition arrays.

## V. CONCLUSION

Good agreement between our experimental results and those obtained by Popović *et al.*<sup>6</sup> in pulsed arc plasma proves the reliability or our measurements since their results represent the most accurate experimental results available at this time (according to Konjević *et al.*<sup>16,17</sup>).

The linear discharge used in this experiment was found to be a very reproducible nitrogen plasma source.

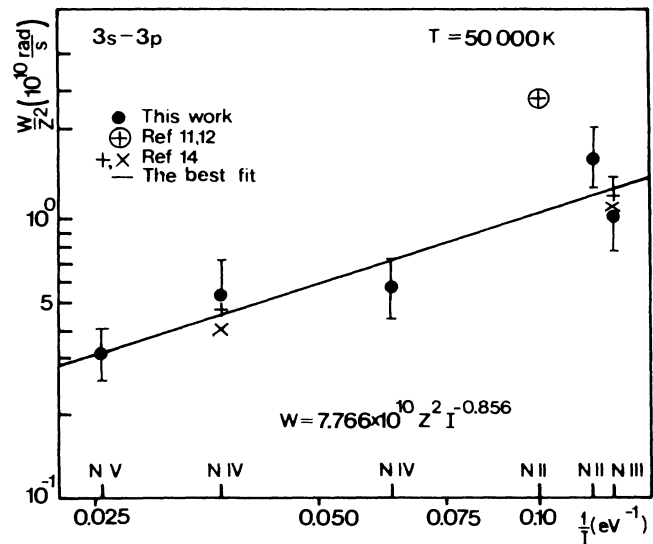


FIG. 6. Stark HWHM ( $w/Z^2$ ) vs inverse value of the upper-level ionization potential ( $I$ ) for N II, N III, N IV, and N V  $3s-3p$  transition arrays of electron temperature  $T = 50\,000$  K and electron density  $N = 10^{23}$  m<sup>-3</sup>: ●, our experimental results; ⊕, the theoretical results of Griem and co-workers (Refs. 11 and 12); + and ×, modified semiclassical and semiempirical results, respectively, taken from Ref. 14.  $Z = 2, 3, 4,$  and  $5$  for N II, N III, N IV, and N V, respectively.

The measured Stark-width results were sufficiently accurate (within  $\pm 15\%$ ) to be used for determining the possible regularities within the investigated transition arrays within one stage of ionization or among different stages of ionization. They verify Eqs. (1)–(3) with an accuracy better than 30%. This is as good as the overall accuracy of the discussed theoretical approaches: semiclassical<sup>11,12</sup> (in the case of N II spectral lines), modified semiclassical,<sup>13-15</sup> and modified semiempirical<sup>13-15</sup> (in the case of N III and N IV spectral lines). Therefore, Eqs. (1)–(3) can be used for the theoretical prediction of Stark-width data for uninvestigated spectral lines, but belonging to N III  $3s-3p$  and  $3p-3d$  transition arrays separately; and to N II, N III, N IV, and N V  $3s-3p$  arrays treated altogether. In both cases the same accuracy has been found up to  $\pm 30\%$ , which is as good as the observed agreement among different experiments.<sup>1-10</sup>

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