Resolving an anomaly between measured spectral linewidths of $n=3$ transitions in N II and O III spectra

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A reported anomaly in the experimental scaling of the widths of Stark broadened $n=3$, $\Delta n=0$ spectral lines along the carbon isoelectronic sequence is not observed in the present experiments. The ratio ω_N/ω_O of widths for N II lines compared to those for the narrower O III lines for the same transitions is now measured as lying between two theoretical predictions, both of which show a value ω_{z-1}/ω_z greater than unity continuing throughout the sequence. In the earlier measurements, the widths (in frequency units) were actually measured as being smaller for N II than for O III , i.e., a ratio ω_{z-1}/ω_z less than unity.

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I. INTRODUCTION

There currently exists considerable interest in the Stark broadening of spectral lines originating on $\Delta n=0$ transitions such as 2*s*-2*p* or 3*s*-3*p* in low- to medium-weight elements, where there is evidence of disagreement between theory and plasma experiments. Factors of two or more discrepancies in measured linewidths occur as the atomic number varies along an isoelectronic sequence [1]. One such experimental scaling study using a pulsed arc involved $n=3$ transitions in carbonlike ions, including N II [2] and O III [3]. In a graph [3,4], which also includes F IV and Ne V, a steady increase in widths for a decreasing charge from Ne V to O III was followed by a surprising decrease for N II , to the extent that the widths (in frequency units) actually became less than those of O III. This has been compared to theoretical estimates [5,6] which predict a significant increase from O III to N II, rather than a decrease. The present experiment has been designed to test this apparent anomaly.

II. EXPERIMENT

The present N II and O III measurements were carried out using a plasma created in a puff of air initially at a pressure of 4 atm, passing through a 1-mm-diameter nozzle, and following a fast-opening valve. Helium was added to the fill gas at a concentration of 0.6% to provide supporting measurements of electron density and temperature from He II and He I linewidths and from He II: He I integrated-line-intensity ratios, respectively. As shown in Fig. 1, the puff was irradiated with a ruby laser operating at a wavelength of 694 nm and nominally 3 J of energy in a 20-ns pulse. The laser beam was focused to a 180-μm-diameter spot at a distance from the nozzle of 160 μm, for an irradiance of 7.5×10^{11} W/cm^2. This arrangement has proven to produce quite a uniform plasma [7–9].

As shown in Fig. 1, radiation from the plasma was focused along an axis orthogonal to both those of the laser beam and the axis of gas expansion onto the 100-μm-wide entrance slit of a 0.75-m stigmatic Czerny-Turner configured spectrograph for use between 320 and 590 nm wavelength. At the exit plane was attached a charge-coupled device (CCD) detector which provided on each shot spectral as well as some spatial resolution along the direction of gas expansion from the nozzle. With a 100-μm-wide entrance slit, a spectral resolution of 0.1 nm was obtained for the N II lines and 0.05 nm for the O III lines, the latter in second order. Such instrumental widths were much less than the typical measured linewidths from the plasma, which ranged from 1 to 2.5 nm at the high electron densities present.

To obtain some limited spatial resolution, the CCD output was binned along the spectral-line direction into ten 1.1-mm-wide segments. Contour maps of intensity covering all ten bins were obtained with total emission passing through the spectrograph at zero order, an example of which is shown in Fig. 2. Some displacement of the plasma along the laser axis at increasing distance from the nozzle was associated with the laser focal position relative to the nozzle port, such that the focal position could be optimized by minimizing this displacement of the image.

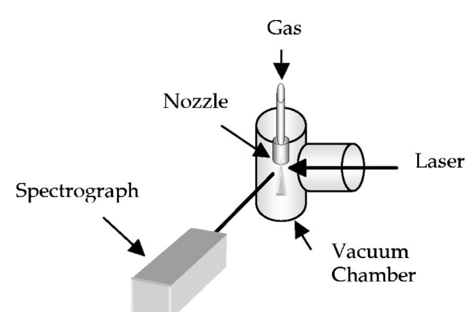

FIG. 1. The layout of the experiment.

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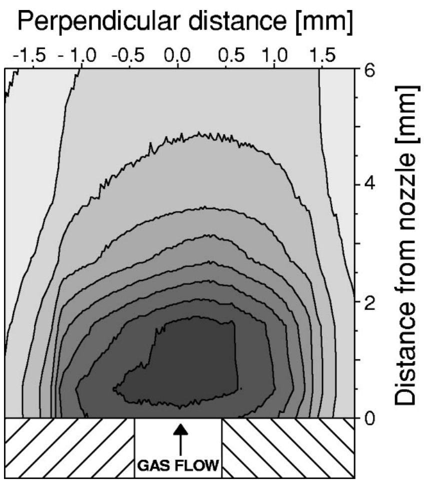

FIG. 2. Time-integrated contour map of the total plasma emission. Also indicated is a section of the nozzle, showing the 1-mm-diameter opening for gas flow. The various contours correspond to changes in steps of 10% in emission, or ~3% in density. The laser was incident from the left and focused at a distance of 160 μm from the nozzle surface. The views for the present spectroscopic measurements were centered at distances of 1.6±0.5 and 6.0±0.5 mm.

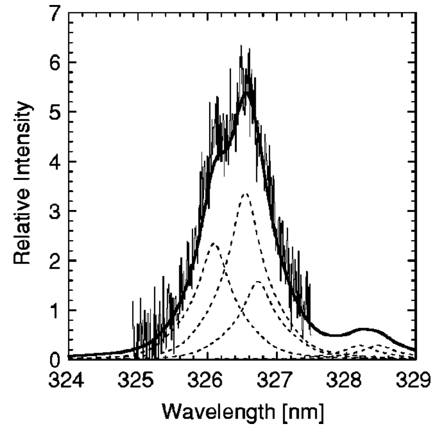

FIG. 3. Comparison of the measured line profile for O III, multiplet no. 102, with a numerical summation of the Lorentzian profiles for the individual lines. A 0.05-nm-wide measured instrumental profile is included as an additional Lorentzian.

III. SPECTROSCOPIC RESULTS

The two multiplets of interest and mean wavelengths [10] are listed in columns 1–3 of Table I. At the high electron density in the present experiment the lines within each multiplet are mostly merged, at least at the highest density in the main portion of the plasma. The overall measured blend of individual line profiles was fitted to calculated sums of Lorentzian intensity distributions, each including a Lorentzian-fitted instrumental profile. A single-layer model for the plasma was assumed. An example of such a fit for O III, multiplet no. 102, is shown in Fig. 3. For these fits, tabulated gf values [10] were used for the relative intensities between components ("g" being the statistical weight of the lower level and "f" the absorption oscillator strength). The resulting ratios $[\omega_N/\omega_O]_{meas}$ of full widths at half maximum (FWHM) in angular frequency units for N II and O III are listed in column 5 of Table I. These measurements fall within the values for the calculations in columns 6 and 7; but they are twice the value from the earlier measurements shown in column 4. The estimated uncertainties in the N II to O III width ratios shown in Table I for the present experiment are ±20%, based on an rms value involving a precision of ±15% in the numerical fitting of the profiles and ±7% in reproducibility between shots with similar laser energies.

One might question whether or not these overlapping lines can simply be represented by a sum of Lorentzian (impact) profiles, suitably centered according to their unperturbed wavelengths, without regard to any individual Stark shifts or, rather, their differences. Possible corrections to this superposition would be forbidden components [11] due to dipole interactions between free electrons and ions. However, the corresponding atomic matrix elements between upper (and lower) levels of the lines are vanishingly small, according to selection rules. Simple superposition is therefore well justified.

The data shown in Table I, column 5, from the present experiment were obtained at a distance of 1.6±0.5 mm from the nozzle surface (at closer distances continuum radiation dominated the spectra). Further measurements were carried out at a distance of 6±0.5 mm from the nozzle surface, where a lower density existed with less line merging. At these distances from the laser focus and at times long after the laser pulse ends, the plasma is in an expanded and relatively quiescent recombination phase (following rapid ionization near the laser focus), such that time-integrated measurements of line ratios between species are reliable.

TABLE I. Comparison of linewidth ratios ω_N/ω_O between N II and O III.

Transition	Wavelength[a] (nm) (multiplet) N II	O III	$[\omega_N/\omega_O]_{meas}$ Refs. [2,3]	$[\omega_N/\omega_O]_{meas}$ Present	$[\omega_N/\omega_O]_{theor}$ SC/SSC[b]	$[\omega_N/\omega_O]_{theor}$ MSE[c]
$3p^3D$-$3d^3F^o$	500.45 (114)	326.57 (102)	0.69±0.07	1.54±0.3	1.90	1.39
$3s^3P^o$-$3p^3D$	567.93 (73)	376.23 (71)	0.83±0.08	1.60±0.3	1.95	1.50

[a]Mean wavelength for the multiplet [10].
[b]SC/SSC: Semiclassical (N)/simplified semiclassical (O) theories [5].
[c]MSE: Modified semiclassical theory [6] as plotted in Refs. [3,4].

The linewidth data [2,3] obtained in the earlier experiments at a relatively low electron density of typically 3×10^{16} cm^{-3} and an electron temperature varying from 1.5 to 3.5 eV gave the average linewidth ratio when averaged over multiplet components shown in column 4 of Table I, which, once again, is much lower than both that expected theoretically and that measured here.

For determining plasma parameters, first the He II $3d$-$4f$ spectral blend at a wavelength of 468.6 nm was fitted with a single Lorentzian distribution to which was added a Lorentzian width characteristic of the instrumental function. This yielded, from Stark broadening calculations [5], an electron density $N_e = 3 \times 10^{18}$ cm^{-3} in the more dense portion of the plasma at a distance of 1.6 mm from the nozzle, and 0.4×10^{18} cm^{-3} at a distance of 6 mm. A similar density was obtained with the He I $2p$-$3d$ spectral blend at a wavelength of 587.5 nm by combining two Lorentzian distributions, corresponding to a two-layer plasma. This was necessary because of a narrow central component from a cooler outer layer for neutral helium. These helium widths are ~20% lower than N II widths obtained using calculated values tabulated in Ref. [5]. (We assume here that the ion-collisional broadening is negligible, as discussed in Refs. [5,12].)

The ratio of the integrated intensity of the He II line to that of the broader component of the fit to the He I line resulted in electron temperatures of $kT_e = 4.8 + 0.5$ eV and 3.9 ± 0.5 eV at the same two respective distances from the nozzle. These values derived first from a near-LTE (local thermodynamic equilibrium) model [11]. They were subsequently supported by numerical modeling using a steady-state collisional-radiative equilibrium (CRE) code [13] which yielded $kT_e = 5.0$ eV and 4.0 eV at the same two positions. Also, this code predicted an electron temperature in the dense region of $kT_e = 4.5$ eV from a measured ratio of O III to N II total line intensities for the multiplets shown in Table I.

IV. CONCLUSION

An unexpected previously observed reversal in the increase in linewidths with decreasing nuclear charge, resulting in a decrease of N II widths below those of O III for the same transitions, is not found in the present experiment. Rather, the presently measured ratios for these two species fall between theoretical predictions, rather than below both theoretical values as was found in the earlier experiments. Two sets of present measurements agree with each other for electron densities differing by a factor of 7. It is suggested that in the previous experiments where a disagreement was found, perhaps nitrogen from a less-dense outer layer (compared to that from which the density is measured using He II linewidths) may have contributed to the illusion of an unusually narrow line.

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