Measurement of the Electron-Impact Broadening of Ionized Nitrogen and Carbon Resonance Lines in a High-Pressure Electric Shock Tube

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Profiles of the overlapping fine-structure components of C II and N II vacuum ultraviolet resonance lines emitted from optically thick shock-heated layers consisting mostly of helium were measured on a shot-to-shot basis, using a T-type electromagnetic shock tube as a source. Plasma conditions were determined from spectroscopic measurements in the visible region. The analysis was based on a radiative transfer model for the resonance multiplets, assuming superimposed Lorentzian profiles with known oscillator strengths and the existence of local thermodynamic equilibrium. The resulting damping constants are interpreted as due mainly to elastic electron collisions and agree within factors of ~1.0 and ~1.5, respectively, with predictions based on a semiempirical method.

I. INTRODUCTION

Agreement between measured[1] and calculated[2] electron-impact broadening of neutral atom lines has, on the whole, been satisfactory (~ ±20%), although the calculations have entailed a large number of approximations, e.g., straight classical perturber paths, long-range dipole interactions, second-order perturbation theory, neglect of lower-state broadening, use of Coulomb-approximation atomic matrix elements, and LS coupling. On the other hand, measured widths of positive ion lines[3] have generally been found to exceed original predictions[2] by factors of 2 to 10. This aroused considerable interest and was followed by a number of more or less involved calculations,[3] all of which have lacked a sound theoretical basis.[4,5] The very simplest of these calculations,[4] based on "optical" cross sections from the effective Gaunt-factor approximation[6] for electric-dipole transitions extrapolated at constant Gaunt factor to zero electron velocity, agreed as well with the available experimental data as did the most detailed quasiclassical calculation,[7] namely, to an average factor of 1.5, with deviations exceeding a factor of 2 as rare exceptions. The effective Gaunt-factor method had been developed for inelastic cross sections, and its extension[4] to energies below threshold, therefore, amounted to a prediction of the contribution from elastic collisions, which cannot be calculated from second-order classical-path perturbation theory.[5]

Because of the lack of experimental data for electron kinetic temperatures (kT_e) much smaller than threshold energies (ΔE) for inelastic dipole-allowed transitions from the levels involved in the transition, the above effective Gaunt-factor extension[4] has had to remain conjecture for the time being. To justify it theoretically, nothing short of a fully quantum-mechanical evaluation of Baranger's general formula[8] in terms of elastic scattering amplitudes and inelastic cross sections for both upper and lower states of the line would do, since for $kT_e \lesssim \Delta E$, the classical-path approximation cannot be justified in the case of positive ion lines.[5] This has recently been accomplished for the resonance lines of singly ionized magnesium[9] and calcium.[10] We set out, simultaneously, to check experimentally the validity of this conjecture for ions with more complicated level structure.

From the beginning, it was clear that the $kT_e \ll \Delta E$ range could be reached only for resonance lines. Unfortunately, their Stark widths are much smaller than Doppler widths under such laboratory conditions where the layers involved are of small optical depth. Therefore, optically thick layers were required to enhance the wings on the expected folded Doppler and Stark profiles. It was unavoidable that for ions such as C II and N II, which are particularly suitable for introduction into plasmas and have $\Delta E \neq 0$, fine-structure components of the resonance multiplets would overlap each other. The measurable quantity was accordingly the composite profile of overlapping lines, the key quantity being the extreme long- and short-wavelength wings resulting from the superposition.

A situation such as this requires considerable analysis if damping constants are to be deduced from measured profiles. Such an analysis would be all but impossible except in a case of an emitting plasma whose properties do not change significantly along the line of sight. Earlier success on a somewhat similar problem, namely the measurement of the Stark broadening of hydrogen resonance lines, encouraged the use of a highly reproducible electrically driven, high-pressure

shock tube on a shot-to-shot scan basis.[11] For a period of several microseconds, this device produces a helium plasma with $kT_e \approx 2$ eV and electron density $N_e \approx 3\times10^{17}$ cm^{-3}, to which small mixtures of test gases may be added. Its usefulness has since been verified by the continued agreement of the measured Lyman-α profiles to within ~± 20% with even the most circumspect calculations.[12–14] Experimental errors in the present case of a blended multiplet could be somewhat larger, but should still allow a meaningful comparison with theoretical predictions.

II. APPARATUS

The electrically driven high-pressure T-type shock tube used was essentially that described in Ref. 11, except that the fused silica used for the arc T section was replaced by aluminum oxide. Also, the capacitive-energy storage system was charged to a potential of 8.7 kV. A shock wave, well separated from any arc-produced contaminated plasma, arrived (after reflection) at an observation position 70 cm from the electrodes and 3.4 mm from the reflector 54 μsec after the discharge. To ensure maximum reproducibility, this total-shock transit time was maintained from shot-to-shot to within ± 0.2 μsec by slight adjustments of the charging voltage.

The gas mixture for the study of the N II resonance-line profile was 40-Torr He, 0.4-Torr H_2, and 0.1-Torr N_2, while that for the C II study was 40-Torr He and 0.2-Torr CH_4. The gases were introduced into a premixing chamber in order of increasing partial pressure. Independence of the data with respect to a varied fill-to-discharge interval indicated sufficient gas mixing prior to discharge.

The helium gas provided the hydrodynamic medium. However, the reflected shock wave was nonluminous unless traces of hydrogen[15–18] (or another gas of low ionization potential) were added to shorten the ionization-relaxation time of the plasma. The amount of minority gas required[17] was found to be ~1% to provide sufficiently short relaxation times to satisfy local thermodynamic equilibrium (LTE) conditions[2,19] for the energy levels of interest.

The gas mixture was admitted into the shock tube at a higher pressure and the capacitor bank was discharged when the pressure in the tube had decreased to the desired level by flow through the (windowless) spectrograph entrance slit into a differentially pumped chamber. The tube pressure at the moment of firing was maintained to ± 5% of the nominal 40 Torr.

The plasma present behind the reflected shock wave was simultaneously observed radially with spectrometers in both the visible and vacuum ultraviolet (N II 1085 Å and C II 1335 Å) spectral regions. The signals were detected by photomultipliers, and in the vacuum-ultraviolet region sodium salicylate was used as a scintillator.

In order to prevent a blocking of the narrow (0.025-mm) vacuum-spectrograph slit by the diffusion of contaminants from the arc region following the event (and stopped short of the observation region during the discharge by the reflected separated shock front[11,17,18]), helium gas was introduced near the front of the vacuum-spectrograph entrance slit and pumped out through one of the shock-tube electrodes. In addition, after about seven discharges it was found necessary to reverse flush this slit by filling the vacuum spectrograph with helium and again pumping through an electrode. This accumulative obstruction of the slit could be continuously monitored optically using a small mirror located between the entrance slit and the grating of the vacuum spectrograph.

The gradual decrease in optical transmission due to deposited material on the tube wall was monitored using a stable light source.

III. AUXILIARY DIAGNOSTICS

An electron density of $N_e = (3.2 \pm 0.3)\times10^{17}$ cm^{-3} was determined for both nitrogen- and carbon-seeded plasmas from a scan of the He I 3889-Å line profile[20] using a monochromator with an instrumental width of 1.3 Å.

An electron temperature of $(19.0 \pm 0.6)\times10^3$ °K was also determined for both test elements from the ratio of the total intensities of the He I 4713-Å and Hβ 4861-Å lines assuming local thermodynamic equilibrium (LTE) to exist, at the measured electron density, between the populations of relevant energy levels. LTE was indicated from calculated excitation-ionization relaxation times[2,19] which are at least an order of magnitude shorter than the observation time (~4 μsec) following the shock arrival. The relative total line intensities were obtained from a scan of the line profiles. The monochromator and photomultiplier used were calibrated for relative wavelength response over the spectral range of interest using a tungsten-ribbon standard lamp.

A further sensitive check on the temperature equivalence for the two cases was obtained from comparing the intensity at the (flat) peaks of the N II multiplet components (associated with the saturation intensity as determined by the blackbody limit at the plasma temperature) for both He-H_2-N_2 and He-CH_4-N_2 (0.025 Torr of N_2) mixtures. The agreement was within 20% (equivalent to 3% in the temperature).

Another monochromator monitored a 20-Å band

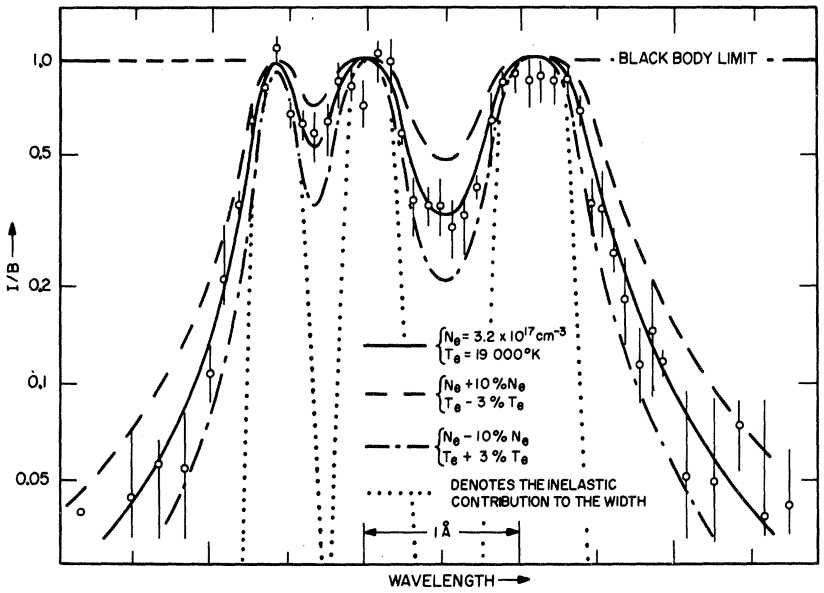

FIG. 1. Comparison of the experimental data points (intensity I relative to the blackbody level B) for the N II resonance multiplet near 1085 Å with a fitted profile (solid line) calculated using an optically thin width corresponding to 1.5 times that expected from theory. Additional theoretical profiles (dashed) corresponding to uncertainties in experimentally determined plasma parameters are shown, as is a profile (dotted) expected if only inelastic and superelastic collisions were present.

of continuum radiation centered at 5423 Å. The primary purpose of this monitor was to enable data analysis at an instant of constant electron density of which visible continuum emission is a sensitive function.[2]

IV. VACUUM ULTRAVIOLET DATA

Figures 1 and 2 present the data for N II and C II, respectively, acquired using a two-meter normal-incidence vacuum-ultraviolet scanning spectrometer with an instrumental width of ~ 0.14 Å. The experimental uncertainties in the data are indicated by flags on the individual points and are seen to lie between theoretical[4] (dashed) curves for the limits of error expected in the measured values of electron density and temperature. The central profile is the theoretical best fit to the data for $N_e = 3.2 \times 10^{17}$ cm^{-3} and $T_e = 19\,000$ °K.

The N II 1085-Å profile was obtained by averaging the data from three discharges at each 0.075-Å step in the wavelength scan, the acceptable discharges being those for which variations in the total-shock transit time were less than 0.2 μsec. The data were read nominally at 4 μsec following the shock-front arrival, the precise time of reading being that at which the electron-density moni-

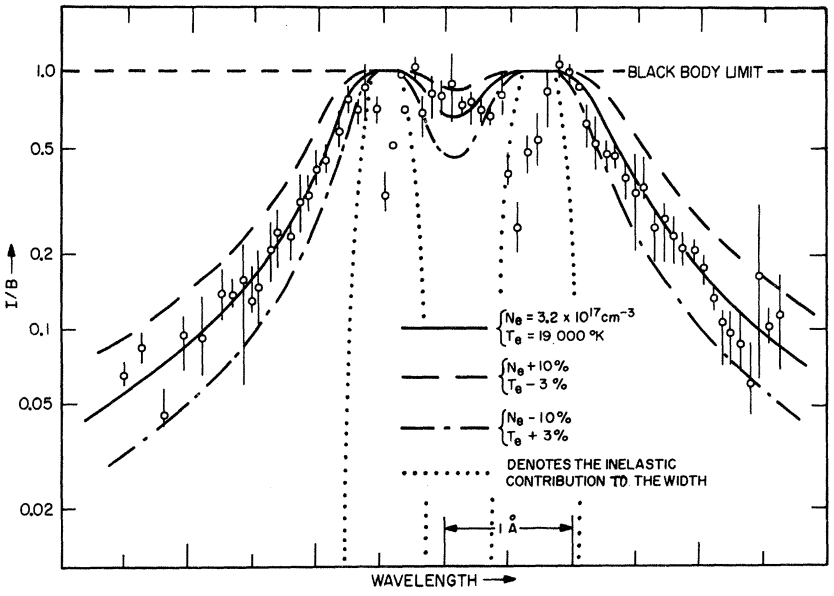

FIG. 2. Comparison of the experimental data points (intensity I relative to the blackbody level B) for the C II resonance multiplet near 1335 Å with a fitted profile (solid line) calculated using an optically thin width corresponding to 1.0 times that expected from theory. Additional theoretical profiles (dashed) corresponding to uncertainties in experimentally determined plasma parameters are shown, as is a profile (dotted) expected if only inelastic and superelastic collisions were present.

tor signal decayed to a predetermined value (subject to a systematic correction for a gradual decline in wall transmission). Over-all systematic errors in the vacuum-ultraviolet scan were determined to be negligible by rescanning the first wing of the profile to the first peak. The C II (1335-Å) profile was obtained similarly.

The optically thick calculated profiles shown in Figs. 1 and 2 were computed[16] using the equation of radiative transfer, assuming a homogeneous layer, and include a triangular instrument function. For this calculation, profiles of the absorption coefficient were assumed using widths varying about recent semiempirically obtained values[4] and using absolute oscillator strengths for the multiplet from recent measurements[21] and tabulations.[22] LTE between the ground-state and the first-excited-state populations of the ion was assumed since, although the spontaneous decay rate is comparable to the collisional deexcitation rate, the line is optically thick. Also radiative decay rates from higher excited states to the first excited state are an order of magnitude lower than those from the first excited state. Excitation and ionization times are short[2,19] compared to an observation time ~ 4 μsec after shock arrival.

V. CONCLUSIONS AND DISCUSSION

The best calculated fits to the experimentally observed profiles of the N II 1085-Å and C II 1335-Å multiplets (Figs. 1 and 2) yield ratios of experimental to calculated dipole widths of 1.5 and 1.0, respectively, which are within the expected precision of present theory[4] (using a Gaunt factor of 0.2). The significance of these results with respect to the theoretical considerations suggested in the introduction are demonstrated by comparison with the additional profiles (dotted) included in Figs. 1 and 2 which were calculated by considering the contribution of electron-impact broadening through inelastic and superelastic dipole interactions only. The experimental data clearly demonstrate wing amplitudes that cannot be supported by these processes alone. Superelastic (dipole) collisions are more important than inelastic dipole collisions and provide only about 3% of the broadening observed.

Higher (than dipole) multipole contributions which are typically $3k\,T/E_H$, or in this case 0.4 times the total dipole contribution, come from mostly elastic collisions.[9] Here E_H is the hydrogen ionization potential. Making this correction, the ratios of experimental to calculated widths become 1.1 and 0.7, respectively.

Other than by electron impacts, van der Waals broadening by the helium atoms would seem to be more important than by other mechanisms, such as resonance broadening, Stark broadening by ions, or interference with perturber radiation.[23] However, the contribution from this effect can be estimated theoretically[2] to stay well within the expected precision of the present semiempirical theory. Therefore, the above experimental evidence indicates the observation of impact broadening of spectral lines of ions dominated by elastic collisions with electrons.

Since submission of this article questions have been raised by Jalufka and Craig[24] concerning possible inhomogeneities in plasmas produced in T-type shock tubes under certain operating conditions, particularly at high ratios (1:1) of test gas (N_2) to carrier gas (He). Since the present device is much larger and operates at a much higher pressure (33×), higher energy (7×), and with an almost negligible admixture of text gas (< 1%), the plasma conditions are entirely different and the shock wave is most likely pressure driven as in diaphragm-type shock tubes (see Ref. 11). Also, the observation of very narrow absorption bands near the line center further justifies the assumption of negligible wing absorption from surrounding boundary layers.

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[1]W. L. Wiese, in *Plasma Diagnostic Techniques*, edited by R. H. Huddlestone and S. L. Leonard (Academic, New York, 1965), Chap. 6, p. 265.

[2]H. R. Griem, *Plasma Spectroscopy* (McGraw-Hill, New York, 1964).

[3]See Ref. 4 for previous experiments and various theoretical explanations.

[4]H. R. Griem, Phys. Rev. 165, 258 (1968).

[5]H. R. Griem, Comments At. Mol. Phys. 1, 27 (1969).

[6]H. Van Regemorter, Astrophys. J. 136, 906 (1962); M. J. Seaton, in *Atomic and Molecular Processes*, edited by D. R. Bates (Academic, New York, 1962), Chap. 11, p. 374.

[7]S. Brechot-Sahal, Astron. Astrophys. J. 1, 91 (1969); 2, 322 (1969).

[8]M. Baranger, Phys. Rev. 112, 855 (1958), Eq. (77c).

[9]O. Bely and H. R. Griem, Phys. Rev. A1, 97 (1970).
[10]K. S. Barnes and G. Peach, Nature 225, 374 (1970).
[11]R. C. Elton and H. R. Griem, Phys. Rev. 135A, 1550 (1964); see also Naval Research Laboratory Report No. 5967, 1963 (unpublished).
[12]H. R. Griem, Phys. Rev. 140, A1140 (1965); 144, 366 (1966).
[13]E. W. Smith, J. Cooper, and C. R. Vidal, Phys. Rev. 185, 140 (1969).
[14]M. E. Bacon, K. Y. Shen, and J. Cooper, Phys. Rev. 188, 50 (1969).
[15]H. Petschek and S. Byron, Ann. Phys. (N.Y.) 1, 270 (1957).
[16]R. Lincke and H. R. Griem, Phys. Rev. 143, 66 (1966).
[17]J. D. E. Fortna, Ph.D. thesis, University of Maryland, 1969 (unpublished); see also Naval Research Laboratory Report No. 6950, 1970 (unpublished).
[18]K. Fukuda, R. Okasaka, and T. Fujimoto, in Proceedings of the Seventh International Shock Tube Symposium, Toronto, Canada, 1969 (unpublished).
[19]H. Griem, Phys. Rev. 131, 1170 (1963).
[20]H. R. Griem, M. Baranger, A. C. Kolb, and G. Oertel, Phys. Rev. 125, 177 (1962).
[21]G. M. Lawrence and B. D. Savage, Phys. Rev. 141, 67 (1966); see also L. Heroux, *ibid.* 153, 156 (1967); S. Bashkin, H. G. Berry, W. S. Bickel, J. Desesquelles, Compt. Rend. 269, 63 (1969); J. Bromander, R. Buchta, and L. Lundin, Phys. Letters 29A, 523 (1969).
[22]W. L. Wiese, M. W. Smith, and B. M. Glennon, *Atomic Transition Probabilities, Vol. I, Hydrogen Through Neon* (U. S. GPO, Washington, D.C., 1966).
[23]D. D. Burgess, Phys. Rev. 176, 150 (1968).
[24]N. W. Jalufka and J. P. Craig, Phys. Rev. A1, 221 (1970).

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Temperature Dependence of Helium-Ion Mobilities*

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The mobilities of the He^+ and He_2^+ ions in helium were measured as a function of E/n and gas temperature for $T = 300$–$76\,^\circ K$. At temperatures below $200\,^\circ K$ and at very low E/n, conversion of the He_2^+ to He_3^+ was observed to take place. Mobilities of He_3^+ were determined at 115 and $76\,^\circ K$. The zero-field mobility of He^+ was found to increase with decreasing temperature and was in excellent agreement with the theoretical predictions of Dickinson. Both the He_2^+ and He_3^+ mobilities decreased with decreasing temperature. The He_2^+ mobilities were all significantly lower than the Langevin polarization limit, and were lower even than the mobilities of the heavier He_3^+ ion.

I. INTRODUCTION

Transport properties of ions in gases have been the subject of experimental and theoretical studies for many years. In the presence of an electric field, positive ions immersed in a gas acquire a preferential drift along the field direction. This ion motion is best characterized by the parameter called the ion mobility, which is defined to be the ratio of ion drift velocity v to the electric field strength E (i.e., $\mu = v/E$). For the conditions of low ion densities and weak fields, the energy distribution of the ions is nearly Maxwellian and the mobility is a constant that ultimately depends on the interaction forces between the ions and neutral atoms. Consequently, ion mobility measurements can provide valuable information on the ion–neutral-scattering processes at near thermal energies. Furthermore, variations of the mobilities with parameters like gas temperature and electric field can provide information on the energy dependence of this scattering.

Helium is perhaps the simplest gas system in which to attempt a detailed understanding of the ion transport. Accordingly, there has been a rather long history of helium mobility studies. It was discovered [1] early in this history that two positive-ion species could be observed in purified He gas at pressures of a few Torr and at room temperature. One of these ions was readily identified as being the atomic ion He^+ by virtue of its rather unique mobility. This ion is subject to a resonant charge transfer with the neutral He atoms, and this interaction provides the determining cross section for the transport of charge in the form of He^+. The early availability of quantum-mechanical calculations of the He^+ mobility[2] provided confirmation of the experimental measurements.

The other ion observed in the early experiments was presumed to be the diatomic He_2^+. However, as more experimental work was reported, there developed a discrepancy concerning its mobility. Whereas the early experiments [1,3,4] attributed a room-temperature mobility of about 20.3 cm^2/V