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SPONTANEOUS-EMISSION LINE SHAPE OF ION LASER TRANSITIONS*

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The noble-gas ion lasers currently represent the most intense continuous sources of coherent radiation in the visible spectrum.[1] An understanding of the frequency characteristics of this important class of laser transitions comparable to that available in the case of neutral transitions[2] is clearly dependent on a knowledge of the physical processes which determine the spontaneous-emission line shape under the conditions in the oscillator. These ion lasers are frequently run with applied dc electric fields sufficient to produce Doppler shifts comparable to the natural widths for the transitions during the mean radiative lifetime of the ion. Consequently, non-Lorentzian line shapes are to be expected for these transitions in the general case. We give below a classical derivation of the actual line shape to be expected from an accelerating, radiating ion and evaluate the errors associated with a Lorentzian approximation to this shape. We also present experimental results on representative ion laser transitions in argon for which accurate radiative lifetime data are available.[3,4] These results show that the Lorentzian approximation is extremely good for typical conditions in the Ar^+ laser, due largely to very important nonradiative sources of phase interruption. The most plausible source of this additional line broadening consists of small-angle Coulomb scattering in ion-ion collisions.

Consider an excited ion formed with initial velocity v_0 in the direction of the field and subject to a mean random phase interruption rate R. This ion travels a distance $v_0t + (eE/2m)t^2$ along the field in time t. The Doppler-shifted resonant frequency seen by a fixed observer looking at the receding ion, averaged over time t, will be $\omega_0[1-(v_0/c)-(eE/2mc)t]$ where ω_0 is the resonant frequency of the stationary ion. It follows that the average time-dependent optical field seen by the observer is of the form

$$\langle E(t)\rangle = E_0 \exp\{i\omega_0[1-(v_0/c)-(eE/2mc)t]t - Rt/2\}. \quad (1)$$

The spectral intensity distribution, $I(\omega) = |E_\omega|^2$, is obtained from the Fourier transform of Eq. (1). This results in

$$I_\omega = (\pi/2\xi)I_0|w(z)|^2, \quad (2)$$

where the function

$$w(z) = \{1 + (2i/\sqrt{\pi})\textstyle\int_0^z e^{t^2}dt\}\exp(-z^2) \quad (3)$$

has been tabulated by Faddeyeva and Terent'ev.[5] In the present case the complex argument is given by

$$z = (1-\Omega)/2\xi^{\frac{1}{2}} + i(1+\Omega)/2\xi^{\frac{1}{2}}, \quad (4)$$

where the dimensionless parameters Ω and ξ are defined by

$$\Omega \equiv 2(\omega-\omega_0')/R \text{ and } \xi \equiv 4\omega_0 eE_{dc}/mcR^2. \quad (5)$$

The exact form of Eq. (2) is shown plotted in

Fig. 1 for several values of ξ. Note that in Eq. (5) the constant Doppler shift (v_0/λ) has been absorbed in the resonant frequency ω_0' and that the full Lorentzian width is $R/2\pi$. It has been shown in the nonaccelerated case that R should be replaced by the sum of the upper- and lower-state phase interruption rates in going from the classical to the quantum mechanical limit.[6] We assume that the same transformation should be made here. The total line shape for an ensemble of ions must of course be obtained by integrating the individual response function in Eq. (2) over the ion velocity distribution.

For $\xi \ll |\Omega|$, Eq. (2) becomes

$$I_\omega \approx I_0(1+\Omega^2)^{-1}[1-4\Omega\xi/(1+\Omega^2)^2-\xi^2(5-38\Omega^2+5\Omega^4)/(1+\Omega^2)^4+O(\xi^3)], \tag{6}$$

where the term outside the bracket represents the normal Lorentzian response for $\omega_0/R \gg 1$. Consequently, the exact line shape in Eq. (2) approaches a Lorentzian with peak response I_0 centered at $\Omega=0$, both for the entire line as $\xi \to 0$ and in the extreme wings of the line for arbitrary ξ. An empirical analysis of the numerical line shape in Eq. (2) for $0 \le \xi \le 1$ showed that the maximum multiplicative correction to the original Lorentzian response over the entire line is roughly $(1+2.3\xi^{1.2})$. Also, the full width of the line at half-maximum is increased roughly by the factor $(1+0.45\xi^{1.4})$ and the peak of the line is shifted by $|\delta\Omega_{max}| \approx 1.3\xi^{0.8}$. Note that the direction of the shift depends on the direction of the field relative to the observer.

The dc electric fields present in the positive column of a continuous argon ion laser are ≈ 4 V/cm (for a 2-mm discharge-tube diameter)[7] and the dominant radiative relaxation rates are typically 10^8 and 5×10^8 sec^{-1} for the upper[3] and lower[4] states, respectively. Consequently, if the phase interruption were entirely from spontaneous radiative decay under these conditions we would expect $\xi \approx 0.14$. In that case, maximum departures $\approx 20\%$ would arise from the Lorentzian form and the full width of the line at half-maximum would be broadened by $\approx 3\%$ from the natural width (≈ 110 Mc/sec).

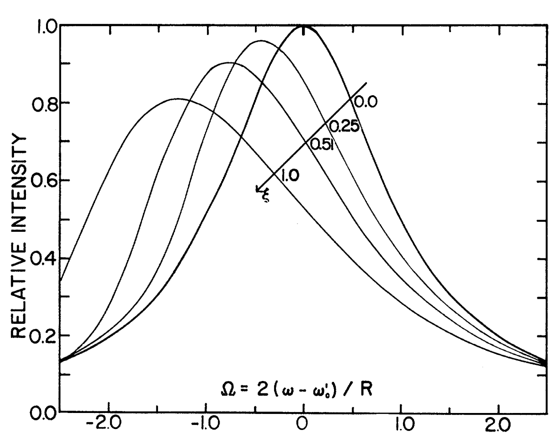

FIG. 1. Classical spectral distribution (I_ω/I_0) from an accelerating, radiating ion as seen by an observer looking in the direction of the field [see Eq. (2)].

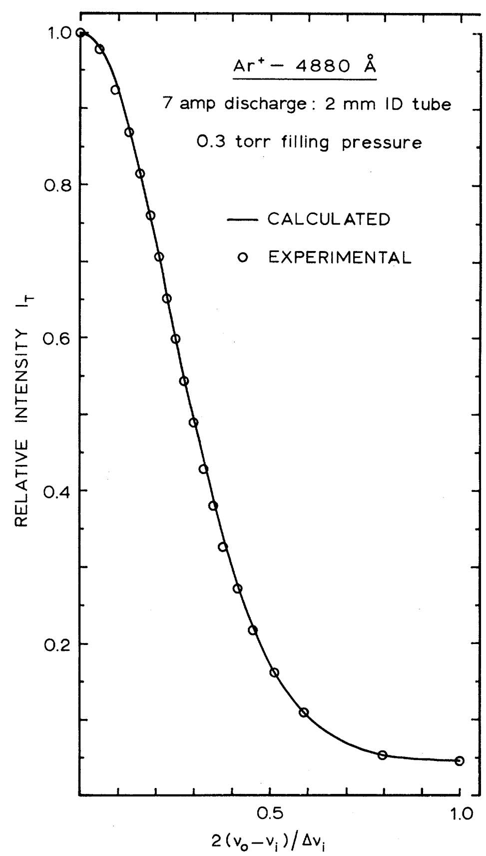

FIG. 2. Comparison of theoretical interferometer response to a Doppler-broadened distribution of Lorentzians with experimental results for a typical Ar^+ laser transition. The theoretical curve was determined using the exact solution described in Ref. 8. The abscissa is proportional to the frequency displacement from the interferometer maximum and is defined in Ref. 8.

Using a method developed by Ballik[8] for analysis of Doppler-broadened Lorentzian line shapes transmitted by a scanning Fabry-Perot interferometer, we have made a study of numerous transitions in argon as a function of various discharge parameters. Detailed results of these measurements will be published later. We wish to note here, however, that the functional form of the observed line shapes of the ion laser transitions corresponds to a Doppler-broadened (Gaussian) distribution of Lorentzians within our limits of instrumental error, <0.5% (see Fig. 2). Further, the Lorentzian widths measured near the optimum conditions for laser oscillation increase with the discharge current and are considerably larger than the characteristic natural widths for these lines. A slight increase in Lorentzian width at filling pressures >1 Torr was also found, which is compatible with phase-interruption widths expected from charge-exchange collisions with neutral Ar atoms. [The resonant ground-state charge-transfer cross sections are $\approx(2 \text{ to } 3)\times 10^{-14}$ cm^2 at the thermal energies in the Ar^+ laser.][9]

It seems probable that the greatly increased Lorentz widths occurring at lower filling pressures (higher ion densities) arise from small-angle ion-ion scattering. If one defines a phase-interrupting collision as one in which the Doppler-shifted resonant frequency of the ion is changed by more than the natural width of the transition, the corresponding cross sections can be calculated directly from the Rutherford formula. The ion densities required by this criterion are somewhat smaller than the electron densities computed from the observed Lorentz widths of neutral argon transitions using the Stark broadening theory of Griem.[10] Griem's theory and previous measurements by Minnhagen[11] indicate that Stark broadening would contribute a negligible amount to the observed Lorentz widths of the ion transitions. Also, calculations based on the known g values for these transitions indicate that Zeeman broadening from the currents in the discharge would represent a small fraction of the total Lorentz widths measured experimentally. A summary of data for the strongest laser transitions is given in Table I. It should be noted that the observed widths ($\approx$500 Mc/sec) at maximum ion density typically correspond to values of the parameter $\xi\approx 0.006$. The latter results in a maximum multiplicative correction to the Lorentzian line shape $\approx$1.005 and an increase over the Lorentz width $\approx$0.2 Mc/sec under the conditions of the experiment. The Lorentzian approximation is therefore extremely good for conditions typical of the argon ion laser. It is to be expected that a similar situation will hold in the other noble-gas ion lasers.

Table I. Observed Lorentz widths at maximum ion density (2-mm capillary at $\approx$0.3-Torr filling pressure). The neutral and ion temperatures under these conditions were $\approx$1400 and 2500°K, respectively.

Line (Å)	Natural width (Mc/sec)	Observed width at 5 A (Mc/sec)
4579	107	500
4658	106	600
4765	107	520
4880	107	500
4965	105	500
5017	···	460
5145	108	680

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[1]For a recent review of gas laser characteristics, see W. R. Bennett, Jr., Appl. Opt. Suppl. 2, 3 (1965).

[2]See W. E. Lamb, Jr., Phys. Rev. 134, A1429 (1964).

[3]W. R. Bennett, Jr., P. J. Kindlmann, G. N. Mercer, and J. Sunderland, Appl. Phys. Letters 5, 158 (1964).

[4]H. Statz, F. A. Horrigan, and S. H. Koozekanani, J. Appl. Phys 36, 2278 (1965).

[5]V. N. Faddeyeva and N. M. Terent'ev, Tables of the Function $w(z)\cdots$ for Complex Argument (Pergamon Press, New York, 1961).

[6]V. Weisskopf and E. Wigner, Z. Physik 63, 54 (1930); 65, 18 (1930); see also W. E. Lamb, Jr., and T. M. Sanders, Phys. Rev. 119, 1901 (1960); W. R. Bennett, Jr., Phys. Rev. 126, 580 (1962), Appendix II; and W. R. Bennett, Jr., Appl. Opt. Suppl. 2, 78 (1965).

[7]A more detailed description of these measurements is to be published by the authors.

[8]E. A. Ballik, Appl. Opt. 5, 170 (1966).

[9]The authors are indebted to Professor Ira Bernstein for the disclosure of his calculated values of ground-state charge-exchange cross sections.

[10]H. R. Griem, Plasma Spectroscopy (McGraw-Hill Book Company, New York, 1964).

[11]L. Minnhagen, Arkiv Fysik 1, 425 (1950).