Asymmetry of Stark-broadened Lyman lines from laser-produced plasmas

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The asymmetries, as well as the widths, of spectral line profiles emitted by highly ionized high-Z elements in a dense, laser-produced hot plasma may be of use in determining the density of these plasmas. Three effects which introduce asymmetries into the line shape are the ion-quadrupole interaction, quadratic Stark effect, and fine-structure splitting. In this paper a method is proposed for including the ion-quadrupole interaction. This technique is then combined with procedures for including the quadratic Stark effect and fine-structure splitting to obtain asymmetric Lyman x-ray line profiles which include all three contributions.

INTRODUCTION AND FORMALISM

The line-shape function, in the static-ion approximation, is given by[1]

$$I(\omega)=\int_0^\infty P(\epsilon)J(\omega,\epsilon)d\epsilon \ , \tag{1}$$

where $P(\epsilon)$ is the static ion microfield probability[2] and

$$J(\omega,\epsilon)=-\frac{1}{\pi}\mathrm{Im}\,\mathrm{Tr}_r D[\omega-(H_r/\hbar-\omega_0)-\epsilon ez/\hbar-M(\omega)]^{-1} \ , \tag{2}$$

which is the electron-broadened line profile for a given ion microfield ϵ. Here we have chosen our z axis to be in the direction of ϵ. The electron-broadening effects are contained in the operator $M(\omega)$,[3,4] Tr_r represents a trace over radiator states, H_r is the free-radiator Hamiltonian, and $\hbar\omega_0$ is the radiator ground-state energy. The operator D is given by $\mathbf{d}\cdot\mathbf{d}$, where the radiator dipole operator is restricted to have nonzero matrix elements only between initial and final states of the Lyman line to be calculated.

This is the general formalism which will be modified in subsequent sections to include asymmetries due to the ion-quadrupole interaction, fine-structure splitting, and the quadratic Stark effect.

ION QUADRUPOLE INTERACTION

Assuming that perturbing ions do not penetrate the radiating ion, the energy of electrostatic interaction between the ions and the radiator can be written as a multipole expansion,[5]

$$U=q\Phi(0)-\mathbf{d}\cdot\mathbf{E}(0)-\tfrac{1}{6}\sum_{i,j}Q_{ij}\frac{\partial E_i(0)}{\partial x_j}+\cdots \ , \tag{3}$$

where q, $\mathbf{d}$, and $\overleftrightarrow{Q}$ are the radiator total charge, dipole moment, and quadrupole tensor, respectively, and $\Phi(0)$, $\mathbf{E}(0)$, and $\partial E_i/\partial x_j$ are the potential, electric field, and electric field gradient at the radiator due to the perturbing ions. This is basically an expansion in a smallness parameter defined as the ratio of the atomic radius and the average interion spacing, $(n^2a_0/z)R_{i0}$, where n is the principle quantum number of the radiator, a_0 is the Bohr radius, Z is the radiator nuclear charge, and R_{i0} is the average distance between ions.[6] For an Ar Ly-α line at an electron density of $10^{23}\ \mathrm{cm}^{-3}$ ($10^{24}\ \mathrm{cm}^{-3}$), this smallness parameter is 0.19 (0.41). In this work, we will terminate the expansion at the quadrupole term. The next-order term, the octupole interaction, is not only 1 order higher in the smallness parameter, but also has, to first order in perturbation theory, a symmetric effect on the line shape, and, hence, should not produce a significant effect on line asymmetry.[7]

The generalization of Eqs. (1) and (2) to include the interaction of the radiator quadrupolar tensor with the electric field gradients of the perturbing ions is given by[8]

$$I(\omega)=\int d\epsilon\int d\epsilon_{xx}\int d\epsilon_{yy}\int d\epsilon_{zz}\int d\epsilon_{xy}\int d\epsilon_{xz}\int d\epsilon_{yz}\,P'(\epsilon,\epsilon_{xx},\ldots)J(\omega,\epsilon,\epsilon_{xx},\ldots) \ , \tag{4}$$

where

$$\epsilon_{ij}=\frac{\partial E_i}{\partial x_j} \ . \tag{5}$$

$P'(\epsilon,\epsilon_{xx},\epsilon_{yy},\epsilon_{zz},\ldots)$ is the joint probability density function for field strength ϵ and field gradients $\epsilon_{xx},\epsilon_{yy},\ldots$. The prime on P indicates that the coordinate frame used is to be one with the z axis along ϵ. $J(\omega,\epsilon,\epsilon_{xx},\ldots)$ is the line shape for given values of $\epsilon,\epsilon_{xx},\ldots$, given by

$$J(\omega,\epsilon,\epsilon_{xx},\ldots)=-\pi^{-1}\mathrm{Im}\,\mathrm{Tr}_r D\times\left[\omega-(H_r/\hbar-\omega_0)-\epsilon ez/\hbar+\frac{1}{6h}\sum_{i,j}Q_{ij}\epsilon_{ij}-M(\omega)\right]^{-1} . \tag{6}$$

The joint probability can be expressed in terms of the usual microfield, $P(\epsilon)$, by writing

$$P'(\epsilon,\epsilon_{xx},\ldots)=P(\epsilon)W(\epsilon\,|\,\epsilon_{xx},\ldots)\;, \tag{7}$$

where $W(\epsilon\,|\,\epsilon_{xx},\ldots)$ is the conditional probability for finding field gradients $\epsilon_{xx},\epsilon_{yy},\ldots$ given that the field has magnitude ϵ and is the z direction.

We define a constrained average $\langle \epsilon_{ij}\rangle_\epsilon$ by

$$\int\int P'(\epsilon,\epsilon_{xx},\epsilon_{yy},\ldots)\epsilon_{ij}d\epsilon_{xx}\,d\epsilon_{yy},\ldots=P(\epsilon)\langle \epsilon_{ij}\rangle_\epsilon\;. \tag{8}$$

We then make the following simplifying approximations:

$$W(\epsilon\,|\,\epsilon_{xx},\epsilon_{yy},\ldots)\rightarrow\delta(\epsilon_{xx}-\langle\epsilon_{xx}\rangle_\epsilon,\epsilon_{yy}-\langle\epsilon_{yy}\rangle_\epsilon,\ldots)\;. \tag{9}$$

That is, the field gradients are replaced by their constrained averages. In the nearest-neighbor limit, expression (9) is exact. We believe that the use of the nearest-neighbor limit for this expression is equivalent to the treatment of the quadrupole interaction given by d'Etat *et al.*[9] In the general case, expression (9) is exact to linear order in $\langle\epsilon_{ij}\rangle_\epsilon$. Although second-order errors which are proportional to $\langle\epsilon_{ij}^2\rangle_\epsilon-\langle\epsilon_{ij}\rangle_\epsilon^2$, are introduced, they should be 3 orders higher in the smallness parameter. Hence we expect the approach presented here to be valid for argon Ly-α and -β lines for electron densities of up to a few times 10^{24} cm^{-3} and for the Ly-γ line up to a few times 10^{23}.

Using the tracelessness of the quadrupole tensor and the symmetry of the x and y directions, we can write

$$\sum_{i,j}Q_{ij}\langle\epsilon_{ij}\rangle_\epsilon=\sum_j Q_{jj}\langle\epsilon_{jj}\rangle_\epsilon =Q_{zz}[\langle\epsilon_{zz}-\tfrac{1}{2}(\epsilon_{xx}+\epsilon_{yy})\rangle_\epsilon]\;. \tag{10}$$

The microfield, $P(\epsilon)$, is calculated for screened fields derivable from a Debye-Huckel form for the potential. To be consistent, the field gradients must also be derived from this potential. The result is

$$\langle\epsilon_{zz}-\tfrac{1}{2}(\epsilon_{xx}+\epsilon_{yy})\rangle_\epsilon =Ze\left\langle\sum_i\frac{e^{-r_i/\lambda}}{r_i^3}(1+r_i/\lambda+r_i^2/3\lambda^2)(1-3\mu^2)\right\rangle_\epsilon\;, \tag{11}$$

where the sum is over perturber ions; r_i and μ_i are the radial coordinate and the cosine of the angle between $\mathbf{r}_i$ and the z axis for the ith ion, respectively.

This constrained average is calculated in the independent perturber model.[10] Within this model the above expression becomes

$$\langle\epsilon_{zz}-\tfrac{1}{2}(\epsilon_{xx}+\epsilon_{yy})\rangle_\epsilon=\frac{-8\pi Zen_i\epsilon\int_0^\infty dk\,k^2e^{nh_1(k)}j_2(k\epsilon)\int_0^\infty dr\,r^2g(r)F(r)j_2(k\epsilon_1(r))}{\int_0^\infty dk\,k\epsilon^{nh_1(k)}\sin(k\epsilon)}\;, \tag{12}$$

where

$$h_1(k)=4\pi\int_0^\infty dr\,r^2e^{-\beta V(r)}[j_0(k\epsilon_1(r))-1]\;,$$

$$F(r)=\frac{e^{-r/\lambda}}{r^3}\left[1+\frac{r}{\lambda}+\frac{1}{3}\frac{r^2}{\lambda^3}\right]\;, \tag{13}$$

$$\epsilon_1(r)=\frac{e^{-r/\lambda}}{r^2}\left[1+\frac{r}{\lambda}\right]\;.$$

n_i is the ion density, j_2 is the second order spherical Bessel function, and $g(r)$ is the radial distribution function. This result is computed numerically. A Debye-Huckel $g(r)$ was used in our calculations. For comparison, a Monte Carlo $g(r)$ was used for a high density case (Ar^{+17} plasma, temperature equals 800 eV, $n_e=10^{24}$ cm^{-3}) with negligible difference in the resulting line shape.

FINE STRUCTURE AND QUADRATIC STARK EFFECT

The fine-structure splitting and the quadratic Stark effect are calculated as in Woltz *et al.*[11] To include fine structure, the Dirac equation is solved for H_r in Eq. (2). The result is diagonal in the representation $|nljj_z\rangle$. It is convenient to expand the exact solution to second order in $(\alpha Z)^2$ with the result,[12]

$$\langle nljj_z\,|\,H_r\,|\,nljj_z\rangle =-\frac{me^4}{2\hbar^2}\frac{Z^2}{n^2}\left[1+\frac{\alpha^2Z^2}{n}\left[\frac{1}{j+\frac{1}{2}}-\frac{3}{4n}\right]\right]\;, \tag{14}$$

where α is the fine-structure constant ($\alpha\approx\frac{1}{137}$). The first term is the usual result when spin is neglected and the second term is the fine-structure correction. The Dirac theory does not contain radiative corrections, but these are of order $\alpha\ln(\alpha)$ smaller than the last term kept above.[12] This result, Eq. (14), is then transformed to the basis $|nlmm_s\rangle$ or $|nqmm_s\rangle$ depending on whether a spherical or parabolic representation is desired. The quantity $[\omega-\omega_0-\epsilon ez/\hbar-M(\omega)]$ is calculated as in the case where spin is neglected;[4] the only difference is that it will have twice as many rows and columns, with the elements being equal to zero if $m_s\neq m_s'$.

We have done two different numerical calculations of Lyman lines in which we have approximately included the quadratic Stark effect. Although the two methods contain quite different approximations they give results which are in close agreement for the Ly-α and -β lines but agree less well for the γ lines. This suggests that while both approximations work well for the α and β lines with the plasma conditions considered here, there is some uncertainty with regard to the Ly-γ calculations.

When the resolvent in Eq. (2) is calculated only within the subspace of initial radiator states (principal quantum

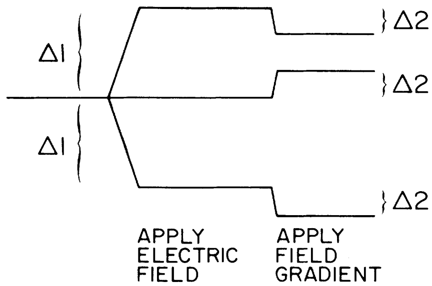

FIG. 1. Energy levels of the $n=2$ hydrogenic argon states in the presence of an electric field and field gradient.

number equals n), the field-dependent radiator Hamiltonian is diagonal in parabolic state representation and its matrix elements give the linear Stark effect

$$E_{nqm}^{(1)} = \langle nqm \mid H_r + \epsilon e z \mid nqm \rangle$$

$$= \left[-\frac{me^4 Z^2}{2\hbar^2 n^2} + \frac{3\hbar^2 \epsilon n q}{2Zme} \right] . \tag{15}$$

Large electric fields, which are present in high density plasmas, can mix states of different principal quantum number and give rise to higher-order Stark effects. A second-order perturbation calculation of the Stark effect gives energy eigenvalues having quadratic field dependence,[12]

$$E_{nqm}^{(2)} = E_{nqm}^{(1)} - \frac{1}{16} a_0^3 \left[\frac{n}{z} \right]^4 \epsilon^2 (17n^2 - 3q^2 - 9m^2 + 19) . \tag{16}$$

The first method of approximately including the quadratic Stark effect consists of calculating the resolvent only within the subspace of initial radiator states, but replacing

TABLE I. Trends in ion-quadrupole interaction, quadratic Stark effect, and fine-structure splitting.

	Principal Quantum Number	Electron Density	Radiator Nuclear Charge
Ion Quadrupole (Blue)	↑	↑	↓
Quadratic Stark (Blue)	↑	↑	↓
Fine Structure (Red)	↓	↓	↑

the linear shift [Eq. (15)] with the quadratic shift [Eq. (16)] for the matrix elements of $H_r + \epsilon e z$. This will give the correct quadratic field dependence, but neglects changes in transition probabilities due to the mixing of states.

The second method is based upon the idea that states with principal quantum number n will mix most strongly with those states which are closest in energy, that is the states with principal quantum number $n+1$. The resolvent matrix is therefore calculated within the subspace of states having principal quantum number either n or $n+1$. This approximation includes the mixing of states with principal quantum number n and $n+1$ but neglects the effects of mixing with other states such as those with principal quantum number $n+2$. Clearly, when Stark shifts are large enough to cause states with principal quantum number $n+1$ and $n+2$ to overlap, it no longer makes sense to neglect this mixing. The condition that $n+1$ and $n+2$ states not overlap puts an upper bound on the electron density for which this approximation is valid. Hence, for Argon plasmas the bound is approximately 2×10^{25}, 2×10^{24}, and 5×10^{23} cm^{-3} for Ly-α, Ly-β, and Ly-γ lines, respectively.

DISCUSSION AND RESULTS

To understand qualitatively the asymmetry due to the ion-quadrupole interaction, we first consider the perturba-

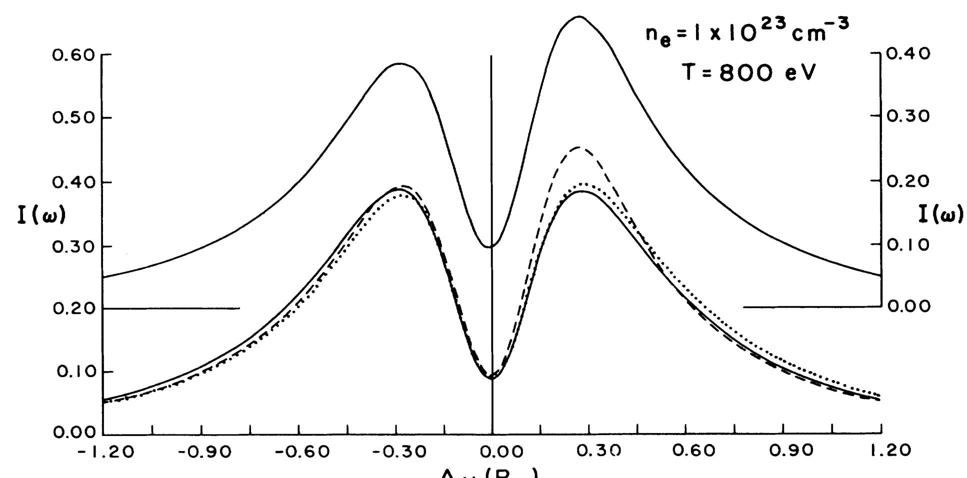

FIG. 2. Argon Ly-β profiles, at an electron density of 10^{23} cm^{-3}, with fine-structure splitting (solid curve), quadratic Stark effect (dashed curve), and ion-quadrupole interaction (dotted curve). Also shown, on a raised scale, is the profile resulting from all three asymmetry considerations.

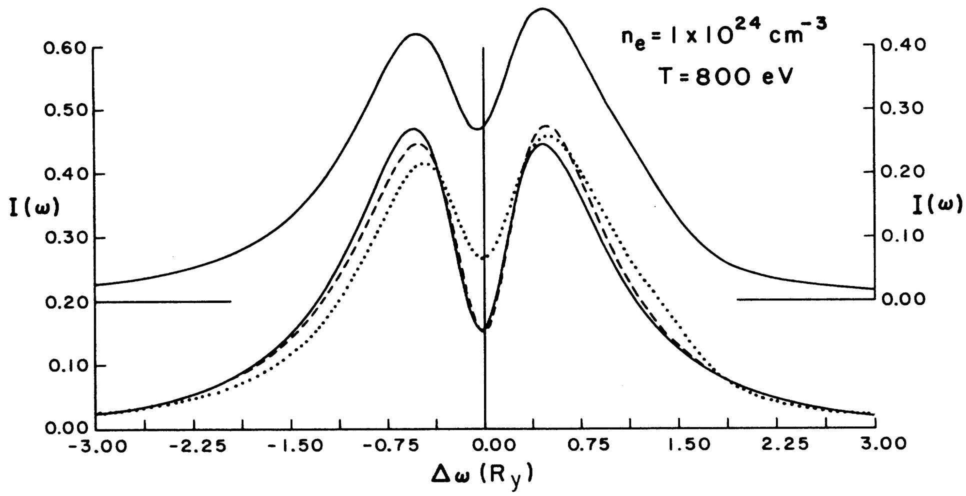

FIG. 3. Same as Fig. 2, but electron density is 10^{24} cm^{-3}.

tion by a single ion located at the coordinates $(0,0,-z)$ on the $n=2$ levels of a hydrogenic ion located at the origin. If we consider initially that the perturbation consists only of the electric field produced by the ion, the new states are, to first order, the well-known Stark states with perturbing field $\epsilon = e/z^2$. If we next include the perturbation due to the field gradients of the ion, the result is as shown in Fig. 1. The higher energy level (which gives rise to the blue wing intensity) is brought in closer to line center while the lower energy level (red wing) is spread further out. This level behavior will lead to a blue asymmetry, at least one to the wings. Similar arguments hold for higher series lines. By considering the smallness parameter,

$$(n^2 a_0/Z)/R_{i0} \sim n^2 n_e^{1/3}/Z^{4/3} ,$$

it is clear that the blue asymmetry caused by the ion-quadrupole interaction will increase with principal quantum number and with density, but will decrease with radiator nuclear charge.

Reference 11 discusses the quadratic Stark effect and fine-structure splitting in more detail. It is shown that the quadratic Stark effect, like the ion-quadrupole interaction, gives rise to a blue asymmetry which increases with principal quantum number and with density but decrease with increasing radiator nuclear charge. The fine-structure splitting has just the opposite effect. It gives rise to a red asymmetry, the magnitude of which decreases with increasing principal quantum number and density while increasing with radiator nuclear charge.

These trends are summarized in Table I.

Figures 2–6 demonstrate these trends. Figure 2 shows argon L-β profiles at an electron density of 10^{23} cm^{-3} and a temperature of 800 eV. Each asymmetry effect is shown separately, and the combined effect is displayed on a raised scale. Density dependence can be seen by com-

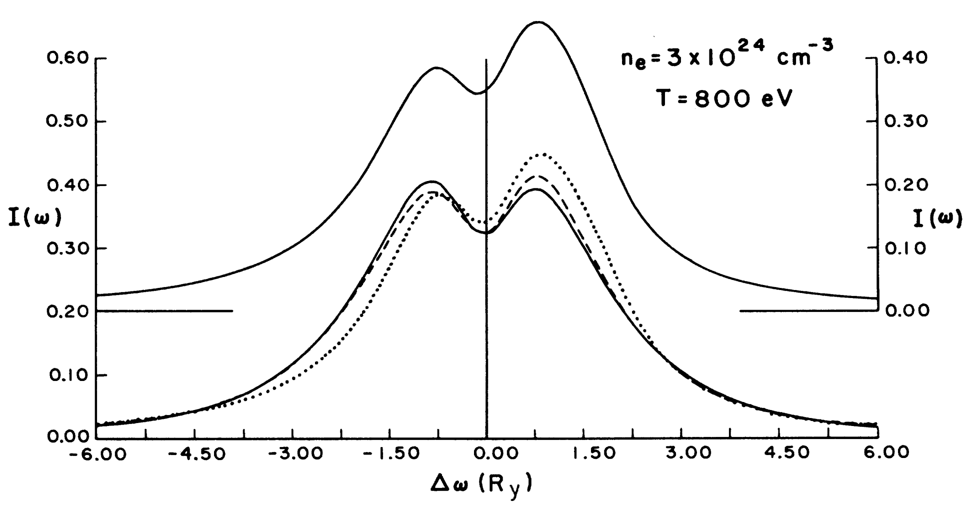

FIG. 4. Same as Fig. 2, but electron density is 3×10^{24} cm^{-3}.

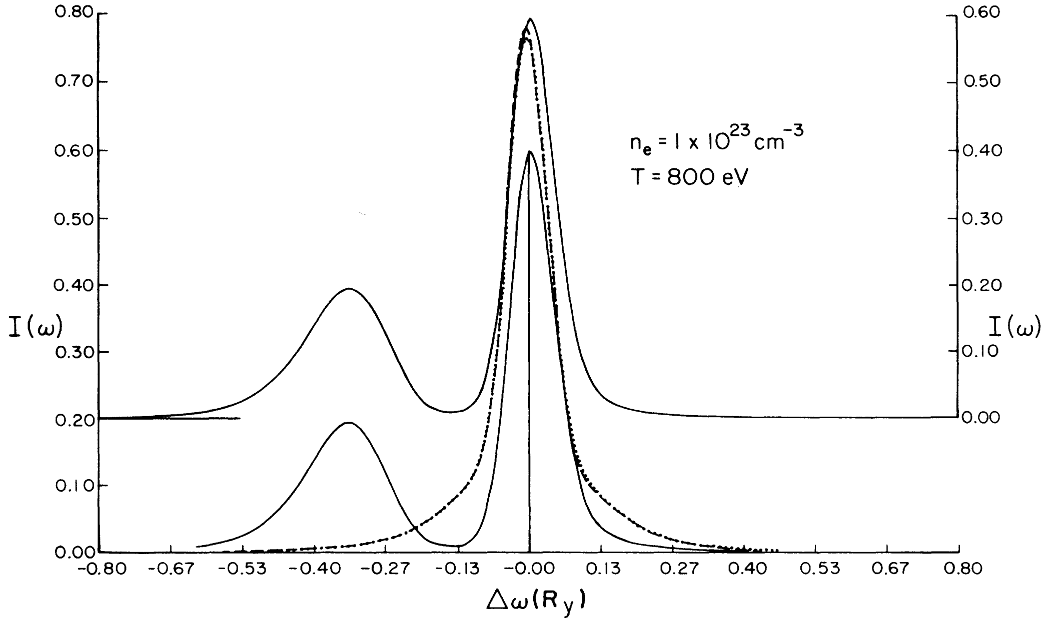

FIG. 5. Argon Ly-α profiles, same conditions as Fig. 2.

paring this with Figs. 3 and 4 which show the corresponding profiles at 10^{24} cm^{-3} and 3×10^{24} cm^{-3}. Figures 5 and 6 show argon Ly-α profiles at densities of 10^{23} cm^{-3} and 10^{24} cm^{-3}. (The peak of one of the profiles in Fig. 6 has been truncated to fit on the figure.) Comparison with Figs. 2 and 3 show the expected principal-quantum-number dependence.

Ly-β lines are particularly useful for density diagnostics because they are usually intense enough to be seen when the Ly-α line is observable, but are not as optically thick. Although inclusion of all three asymmetry effects will cause significant changes in the Lyman line profiles, for most temperature and density regimes, generally the Ly-β line will still be a two-peaked function. Let us define a

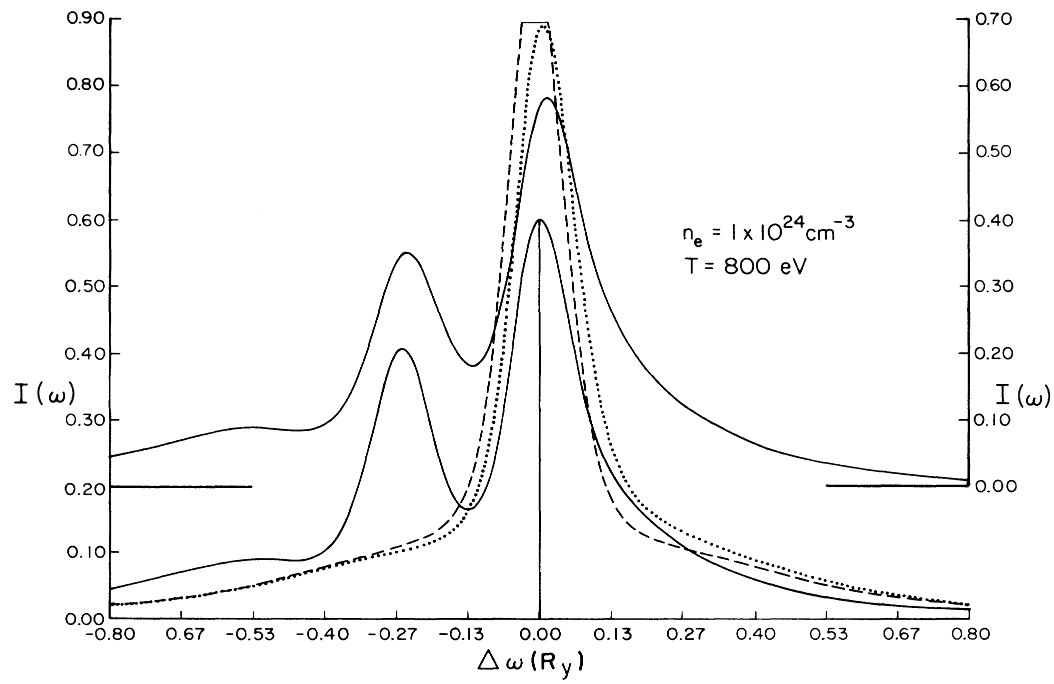

FIG. 6. Argon Ly-α profiles, same conditions as Fig. 3.

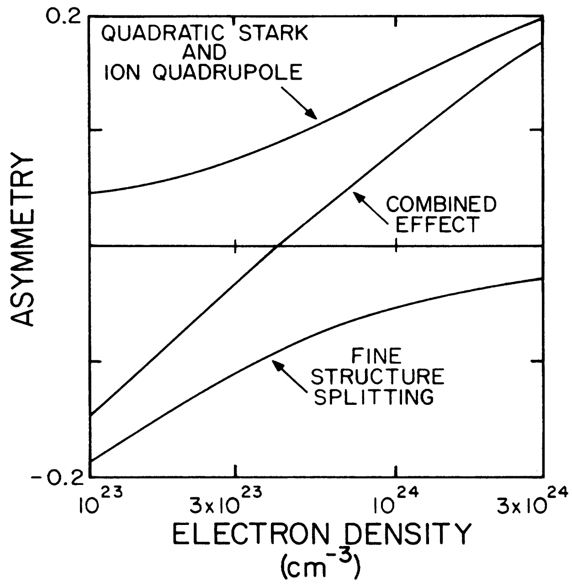

FIG. 7. Density dependence of argon Ly-β line asymmetries due to the fine-structure splitting, the ion-quadrupole interaction, and quadratic Stark effect, and due to the combination of these.

measure of asymmetry of the Ly-β line as $2(I_b - I_r)/(I_b + I_r)$, where I_b and I_r are the blue and red peak intensities, respectively. In Fig. 7, we plot this asymmetry as a function of density for an Ar^{+17} plasma at 800 eV. Lines which include only the fine-structure splitting have a negative (red) asymmetry which decreases in magnitude with increasing density. Both the quadratic Stark effect and the ion-quadrupole interaction cause positive (blue) asymmetries which increase with increasing density. These two effects are shown together in Fig. 7. Also shown is the asymmetry of lines which are calculated using all three effects. It is interesting to note that while each effect, taken alone, would cause significant asymmetry at 4×10^{23} cm^{-3}, their combined effect is to yield a nearly symmetric line profile.

CONCLUSION

A line-broadening theory has been presented which includes three sources of asymmetry, the ion-quadrupole interaction, fine-structure splitting, and the quadratic Stark effect. The behavior of each effect with changes in principal quantum number, electron density, and radiator nuclear charge has been investigated. Of course, when making comparison with experiment, other effects, such as opacity, continuum radiation, and overlapping spectral lines, must be accounted for in the analysis. Nevertheless the asymmetries presented here may be observable in experiments on argon Ly-α and Ly-β lines.[13] However, the noticeable asymmetry of the Ly-γ line does not appear to be in evidence. This may be due to the fact that the Ly-γ lines are emitted under different environmental conditions[14] (i.e., lower average ion density) than the α and β lines or there may be something about the theoretical model that is incorrect for this particular transition. Further theoretical and experimental work is required to resolve these issues.

ACKNOWLEDGMENT

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