

Shifts of spectral lines in a plasma

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The theory of spectral lines in a plasma with quasistatic ions is reconsidered. The ion microfield distribution, generalized to include field gradients and higher-order derivatives, is introduced without decoupling the ion and electron subsystems. It is shown that in a self-consistent treatment of ion-electron correlations the calculations of line shifts requires a more complete theory than has been used in the past. It is shown that level shifts due to a time average of the radiator-plasma interaction are closely related to shifts due to ion field inhomogeneities at the radiator. These two shifting mechanisms are not independent and the analysis indicates how to avoid overcounting. As a result, the time average of the radiator-plasma interaction, usually associated with the so-called plasma polarization shift, is formally different from the equivalent expression given by theories which treat electron-ion correlations in an *ad hoc* fashion.

I. INTRODUCTION

The possibility that a plasma can cause a shift of spectral lines has been a long-standing topic of controversy.¹⁻⁸ The line shift is of fundamental interest since it involves many-body aspects of the line-shape theory due to initial correlations between the radiator and the plasma. It is also of practical interest because, for example, it may be responsible for uncertainties in wavelength determinations and it is a possible diagnostic tool in high compression laser-fusion experiments.

One proposed shift mechanism, first introduced by Berg *et al.*,⁷ is the level shifts due to a nonuniform charge distribution in the neighborhood of the radiator. These authors suggested that a time average of the radiator-plasma interaction can cause an excess negative charge in the vicinity of the radiator. As a result, this net polarization of the plasma partially screens the nuclear charge which alters the level structure of the radiator and shows up as a line shift. In addition, most current theories simplify the difficult problem of predicting a line shape by assuming the quasistatic approximation for the ions and a collisional formulation for the electrons. The static ion approximation implicitly contains a shift due to the ion field inhomogeneity at the radiator.⁹ This second shifting mechanism is in a sense related to the level shifts above in that both involve static perturbations in the potential seen by the bound electrons. Since each mechanism is generally treated independently, there exist the possibility of overcounting some effects.

Also, the usual introduction of the ion microfield distribution assumes a decoupling of the electron and ion subsystems so that electron-ion correlations are treated in an *ad hoc* fashion. There are conceptual difficulties with such procedures. For example, it is empirically observed that the ion microfield distribution should be that for shielded ions where the screening is due to electrons as a result of electron-ion correlations. At the same time, it is known that electron-electron correlations are important in the description of electron-radiator collisions, but no ac-

count is taken of electron-ion correlations. Clearly the *ad hoc* separation of electrons and ions into independent subsystems is difficult to justify theoretically.

Recently a line broadening theory was proposed¹⁰ which retains the quasistatic ion approximation, but treats all plasma interactions (ion-ion, ion-electron, electron-electron) systematically. The approach demonstrated how a self-consistent treatment of ion-electron correlations can be maintained between the ion microfield and the electron collision operator. The discussion in Ref. 10 was restricted to the dipole interaction between the radiator and plasma. Here, the objective is to extend the formalism to a full-Coulomb radiator-plasma interaction. It is then possible to investigate the line shift in a theory which treats plasma correlations self-consistently. The result of the formal analysis shows that the two mechanisms for static line shifts mentioned above, which current theories treat independently, are actually closely related. Moreover, as a direct result of the self-consistent treatment of plasma correlations, the shifts due to the time average of the radiator-plasma interaction are formally different from the equivalent expressions given by the usual theories.

It is important to note that the level shifts due to the time average of the radiator-plasma interaction and ion field inhomogeneity only include static plasma effects due to initial correlations. In order to compare theoretical and experimental shifts, the former must include fluctuations of the time averaged quantities. These fluctuations can be significant since they occur in a time scale much shorter than radiator state lifetimes. Here, however, the discussion is restricted to the static shifts only.

II. THEORY

The line-shape function is determined from the dipole autocorrelation function,¹¹

$$I(\omega) = \pi^{-1} \text{Re} \int_0^{\infty} dt e^{i\omega t} \text{Tr}(\vec{d} \cdot \rho e^{-iLt} \vec{d}), \quad (2.1)$$

where L and ρ are the Liouville and equilibrium density matrix operators for the radiator-plasma system, respec-

tively. The trace is over states of the radiator and plasma. In the following it is assumed that the ions are essentially static over the radiator state lifetimes. Furthermore, it is assumed that the momentum transfer to the radiator during the time of radiation is negligible, and therefore, that the Doppler broadening is independent of Stark broadening. In this situation $I(\omega)$ refers only to Stark broadening so that the operators and trace appearing in Eq. (2.1) are independent of the center-of-mass coordinates of the radiator. It is, therefore, convenient to place the origin of the coordinate system at the radiator nucleus.

To proceed, consider the full-Coulomb radiator-plasma interaction, V_{ap} , which is given by a sum of pairwise additive terms. For simplicity, only a two-component plasma is considered and the radiator is assumed to be hydrogenic with nuclear charge Ze , e the magnitude of the elementary charge. Then,

$$V_{ap} = \sum_{\sigma} \sum_{j=1}^{N_{\sigma}} v^{\sigma}(a, j) \\ = \sum_{\sigma} \sum_{j=1}^{N_{\sigma}} \left[\frac{Z_{\sigma} Z e^2}{|\vec{r}_{j\sigma}|} - \frac{Z_{\sigma} e^2}{|\vec{r}_a - \vec{r}_{j\sigma}|} \right], \quad (2.2)$$

\vec{r}_a and $\vec{r}_{j\sigma}$ are the position operators for the bound electron and j th perturber of species σ , respectively, N_{σ} and $Z_{\sigma}e$ are the number and charge of perturbers belonging to species σ , and σ denotes ion or electron. The pair interaction can be expanded in Legendre polynomials,¹²

$$v^{\sigma}(a, j) = -Z_{\sigma}e \sum_{l=0}^{\infty} \left[\frac{r_{<}^l}{r_{>}^{l+1}} - \frac{Z}{r_{j\sigma}} \delta_{l,0} \right] P_l(\cos\theta). \quad (2.3)$$

In Eq. (2.3) $r_{<}$ ($r_{>}$) is the smaller (larger) of $r_a = |\vec{r}_a|$ and $r_j = |\vec{r}_j|$, θ the angle between \vec{r}_a and \vec{r}_j , and P_l the Legendre polynomial of order l . The terms in the expansion are now regrouped in the following manner:

$$v^{\sigma}(a, j) = \varphi_0^{\sigma}(j) + v_1^{\sigma}(a, j) + v_2^{\sigma}(a, j), \quad (2.4)$$

where

$$\varphi_0^{\sigma}(j) = Z_{\sigma}(Z-1)e^2/r_{j\sigma}, \quad (2.5)$$

$$v_1^{\sigma}(a, j) = -Z_{\sigma}e^2 \sum_{l=1}^{\infty} \frac{r_a^l}{r_{j\sigma}^{l+1}} P_l(\cos\theta), \quad (2.6)$$

for all $r_{j\sigma} \geq 0$ and

$$v_2^{\sigma}(a, j) = \begin{cases} -Z_{\sigma}e^2 \sum_{l=0}^{\infty} \left[\frac{r_{j\sigma}^l}{r_a^{l+1}} - \frac{r_a^l}{r_{j\sigma}^{l+1}} \right] P_l(\cos\theta), & r_{j\sigma} \leq r_a \\ 0, & r_{j\sigma} > r_a. \end{cases} \quad (2.7)$$

It is important to note that the separation of $v^{\sigma}(a, j)$ in

$$I(\omega) = \int d\Psi W(\Psi) J(\omega, \Psi), \quad (2.10)$$

$$J(\omega, \Psi) = -\pi^{-1} \text{Im Tr}_a \{ \vec{d} \cdot f(a; \Psi) [\omega - L(a; \Psi) - B(\Psi) - H(\omega; \Psi)]^{-1} \vec{d} \}, \quad (2.11)$$

Eq. (2.4) is not into contributions from $r_j > r_a$ and $r_j < r_a$. Instead, the different contributions to $v^{\sigma}(a, j)$ are such that φ_0^{σ} and v_1^{σ} are nonzero for all $r_j \geq 0$ and v_2^{σ} is nonzero for $r_j \leq r_a$. The motivation here is twofold. Firstly, the long-ranged monopole term φ_0^{σ} only depends on perturber coordinates and is included in the plasma Hamiltonian as an external potential. In this way the polarization of the plasma by φ_0^{σ} may be accounted for while keeping the rest of the radiator-plasma interaction, v_1^{σ} and v_2^{σ} , short ranged. Secondly, the form of v_1^{σ} is such that it may be written as the "product" of a radiator operator times an operator which depends only on the perturber position,

$$v_1^{\sigma}(a, j) = M(a) \Phi^{\sigma}(j) \\ = - \sum_{k=1}^{N_{\sigma}} \mu_k(a) \phi_k^{\sigma}(j) \\ = - \vec{d} \cdot \vec{\epsilon}^{\sigma}(j) - \frac{1}{6} \sum_m \sum_n Q_{mn} \partial_m \epsilon_n^{\sigma}(j) + \dots, \quad (2.8)$$

where the ellipsis represents higher multipoles. Equation (2.8) defines the expression $M(a) \Phi^{\sigma}(j)$. For example, $\mu_1(a) = \vec{d}$ and $\mu_2(a) = \vec{Q}/6$ are the dipole and quadrupole operators for the radiator, respectively, and $\varphi_1^{\sigma}(j) = \vec{\epsilon}^{\sigma}(j)$ and $\varphi_2^{\sigma}(j) = \partial \vec{\epsilon}^{\sigma}(j)$ are the electric field and field gradient tensor at the origin due to the j th perturber of species σ . The advantage in the product form of $v_1^{\sigma}(a, j)$ in Eq. (2.8) will become apparent later [see Eqs. (2.14) and (2.19)]. Finally, note that in line broadening theories that neglect penetration of the radiator by the perturbers, the radiator-plasma interaction is approximated by $\varphi_0^{\sigma} + v_1^{\sigma}(a, j)$ and the l sum in Eq. (2.6) is usually truncated keeping only the $l=1$ dipole interaction and sometimes the $l=2$ quadrupole interaction.

Now, introduce an ion "configuration" distribution, which generalizes the electric microfield distribution and includes field gradients and all higher-order field derivatives, by defining

$$\rho(\Psi) = \rho \delta(\Psi - \Phi^*) / W(\Psi), \\ W(\Psi) = \text{Tr} \rho \delta(\Psi - \Phi^*), \quad (2.9) \\ \delta(\Psi - \Phi^*) = \prod_{k=1}^{\infty} \delta(\Psi_k - \Phi_k^*),$$

and Ψ constant. The Ψ_k 's and Φ_k^* 's have a meaning similar to ϕ_k^{σ} in Eq. (2.8): for $k=1$ they denote the electric field at the origin, for $k=2$ the field gradient tensor, and so on. The function $W(\Psi)$ is the ion configuration distribution and has the interpretation of a probability density for Φ^* to take the value of Ψ . The form of Φ^* is arbitrary other than depending only on ion coordinates.¹³ Similarly, $\rho(\Psi)$ is the equilibrium density matrix with the plasma constrained to have $\Phi^* = \Psi$. With these definitions, the line-shape function may be expressed in the form¹⁰

where $f(a; \Psi)$ is the density matrix for the radiator

$$f(a; \Psi) = \text{Tr}_p \rho(\Psi), \quad (2.12)$$

Tr_a and Tr_p denote traces over the radiator and plasma degrees of freedom, respectively, and $L(a; \Psi)$ is the Liouville operator for the radiator in the external potential $M(a)\Psi$. The operators $B(\Psi)$ and $H(\omega; \Psi)$ determine the width and shift of the line due to the radiator-plasma interactions not included in $L(a; \Psi)$. The static shift operator $B(\Psi)$ is given by

$$B(\Psi) = \langle L_I(\Phi^*) \rangle, \\ \langle (\dots) \rangle = f^{-1}(a; \Psi) \text{Tr}_p [\rho(\Psi) (\dots)], \quad (2.13)$$

and

$$L_I(\Phi^*) = L_1(\Phi^*) + L_2,$$

where $L_1(\Phi^*)$ and L_2 are the Liouville operators associated with the interactions

$$V_1(\Phi^*) = M(a) \left[\left[\sum_{\sigma} \sum_{j=1}^{N_{\sigma}} \Phi^{\sigma}(j) \right] - \Phi^* \right], \quad (2.14)$$

$$V_2 = \sum_{\sigma} \sum_{j=1}^{N_{\sigma}} v_2^{\sigma}(a, j). \quad (2.15)$$

The operator $H(\omega; \Psi)$ represents collisional effects that require finite times which contribute to the width and dynamic shift of the line and is given by

$$H(\omega; \Psi) = f^{-1}(a; \Psi) \\ \times \text{Tr}_p [L_I(\Phi^*) \rho(\Psi) (\omega - QLQ)^{-1} QL_I(\Phi^*)], \quad (2.16)$$

where $Q = 1 - P$, and P is the projection operator

$$PX = \langle X \rangle \quad (2.17)$$

for X arbitrary.

In obtaining Eqs. (2.10)–(2.17), use has been made of the quasistatic ion approximation and the product form of $v_1^{\sigma}(a, j)$ in Eq. (2.8). The former implies that the modified density matrix $\rho(\Psi)$ is stationary,

$$e^{-iLt} \rho(\Psi) \simeq \rho(\Psi), \quad (2.18)$$

so that the kinetic energy of the ions plays no role and can be integrated out. As a result, the δ function on the definition (2.9) in combination with Eq. (2.8) allows the ion fields characterizing the radiator-ion interaction $v_1^i(a, l)$ to be written as

$$\sum_{j=1}^{N_i} v_1^i(a, j) = M(a) \left[\left[\sum_{j=1}^{N_i} \Phi^i(j) \right] - \Phi^* \right] + M(a)\Psi. \quad (2.19)$$

The last term in Eq. (2.19) depends only on radiator coordinates and is conveniently combined with the Hamiltonian for the radiator internal degrees of freedom to give the Liouville operator $L(a; \Psi)$ appearing in Eq. (2.11). The term with the square brackets $[\dots]$ is combined with $v_1^{\sigma}(a, j)$ to give the definition of $V_1(\Phi^*)$ in Eq. (2.14).

The results in Eqs. (2.10)–(2.17) are of the usual form in line broadening theories making the quasistatic ion approximation: $J(\omega; \Psi)$ represents broadening in the presence of ion configurations such that $\Phi^* = \Psi$, and $W(\Psi)$ gives the probability density of these configurations. The motion of the electrons is accounted for in the definition of $H(\omega; \Psi)$. However, no approximations on the ion-electron correlations are necessary. In fact, Eq. (2.10) is simply an exact rewrite of Eq. (2.1).

III. THE STATIC LINE SHIFT

Although the formal results for the line-shape function in Eqs. (2.10) and (2.11) are exact [within the approximations already present in Eq. (2.1)], the expressions are formidable. For example, in usual line-shape theories the width and shift operator may be evaluated in the binary collision approximation since for typical experimental plasma conditions it involves strong radiator-electron collisions which are well separated in time. Here, however, there are radiator-ion collisions present in $H(\omega; \Psi)$ and these strong collisions overlap except at very low densities. There is also the constraint $\Phi^* = \Psi$ not present in usual line-shape theories. Nevertheless, some general statements regarding static line shifts are possible.

The total static shift (as a result of the instantaneous force on the radiator at $t=0$ due to initial correlations) is contained in $L(a; \Psi) + B(\Psi)$. The shift in $L(a; \Psi)$ is due to the static ion field inhomogeneity at the radiator while the shift in $B(\Psi)$ contains the level shifts given by a constrained plasma average of the radiator-plasma interaction $L_I(\Phi^*)$. Since Φ^* is arbitrary, the shift due to either $L(a; \Psi)$ or $B(\Psi)$ is not unique and, in fact, it is possible to transfer contributions from one term to the other by simply making different choices for Φ^* .

In addition, there is also a frequency-dependent shift contained in $H(\omega; \Psi)$. It is clear from Eq. (2.16) that $H(\omega; \Psi)$ also depends on the choice of Φ^* , and consequently different choices necessarily imply a corresponding transfer of effects between $W(\Psi)$ and $J(\omega; \Psi)$. Again it is emphasized that comparisons of static shifts to experimentally observed line shifts may not be relevant since the latter may contain significant contributions from fluctuations which occur in a time scale much shorter than radiator state lifetimes. Since a self-consistent treatment of electron-ion correlations has been shown to be important in the description of static shifts, it indicates the necessity of including these same correlations in the discussion of the collision operator. Unfortunately, due to the additional complexity of the present formalism (specially the constraint $\Phi^* = \Psi$) such detail analysis has not been possible at this time.

IV. SAMPLE CALCULATION

The purpose of this section is to help clarify the formal results above with a sample calculation of the static shift operator $B(\Psi)$. To proceed, it is necessary to make a choice of Φ^* . Therefore, let

$$\begin{aligned} \Phi^* &\equiv (\text{Tr}_e(\rho_p))^{-1} \text{Tr}_e \left[\rho_p \sum_{\sigma} \sum_{j=1}^{n_{\sigma}} \phi_k^{\sigma}(j) \right] \\ &= \sum_{j=1}^{N_i} \phi_k^i(j) + (\text{Tr}_e(\rho_p))^{-1} \text{Tr}_e \left[\rho_p \sum_{j=1}^{N_e} \phi_k^e(j) \right], \\ &\quad \text{for } k=1,2,3,\dots, \end{aligned} \quad (4.1)$$

where Tr_e denotes a partial trace over the electron subsystem and ρ_p is the density matrix for the plasma and includes the monopole interaction ϕ_0 . The average quantities defined in (4.1) are the conditional average over the electron subsystem given an ion configuration. Note that in this definition Φ^* is completely independent of the ra-

diator internal states since the average is over ρ_p and not ρ . The resulting ϕ_k^* are given by a sum of ion Coulomb terms plus a contribution from electron terms; the latter vanish if electron-ion correlations are neglected.

Although the choice (4.1) involves hindsight and is designed to emphasize a particular result [see Eq. (4.5)], it is certainly valid and involves no approximations. The arguments for justification of (4.1) are not only formal (Φ^* is arbitrary) but also physically reasonable. For example, (4.1) has the empirical desirable feature that the distribution function, $W(\psi)$, is for electron-screened ions. Furthermore, for weakly coupled plasmas the electron screening is given by a Debye theory and the ϕ_k^* 's in (4.1) reduce¹⁰ to the usual form of past calculations.

Substitution of Eq. (4.1) into (2.13) yields

$$\hbar B(\Psi)\chi(a) = f^{-1}(a; \Psi) \sum_{j=1}^{N_e} \text{Tr}_p \rho(\Psi) [1 - (\text{Tr}_e \rho_p)^{-1} \text{Tr}_e \rho_p] [v_1^e(a, j), \chi(a)] + f^{-1}(a; \Psi) \text{Tr}_p \rho(\Psi) [V_2, \chi(a)], \quad (4.2)$$

where $[\dots, \dots]$ denotes a commutator and $\chi(a)$ is an arbitrary operator in the radiator subspace. The two terms on the right side of Eq. (4.2) are quite different. The first involves a constrained plasma average of the radiator-electron interaction, v_1^e , but no radiator-ion terms. Because of the subtracted average over the electrons, this first term describes shifts due to radiator-plasma initial correlations. Note that the relevant initial correlations are those due to $V_1 + V_2$ and not ϕ_0 since the monopole term is included in ρ_p . The second term involves a constrained average over the radiator-plasma interaction describing penetration of the radiator by both electrons and ions and it does not vanish with the neglect of initial correlations.

The result in Eq. (4.2) follows from (4.1) without ap-

proximations. This calculation will make an approximation usually found in static shift theories. In particular, all initial correlations between the radiator and plasma except those due to the monopole interaction, ϕ_0 , will be neglected. No justification for this approximation is given since the intent here is not to provide "improved" numerical results but to make contact with past static shift theories. That is,

$$\rho \rightarrow \rho_a \rho_p, \quad (4.3)$$

where ρ_a is the density matrix for the isolated radiator. Now, the average over v_1^e in Eq. (4.2) vanishes,

$$f^{-1}(a; \Psi) \text{Tr}_p \rho(\Psi) [1 - (\text{Tr}_e \rho_p)^{-1} \text{Tr}_e \rho_p] v_1^e(a, j) \rightarrow \text{Tr}_p \rho_p(\Psi) [1 - (\text{Tr}_e \rho_p)^{-1} \text{Tr}_e \rho_p] v_1^e(a, j) = 0, \quad (4.4)$$

where $\rho_p(\Psi)$ is defined similarly to $\rho(\Psi)$ but with $V_1 + V_2 = 0$. Consequently,

$$\hbar B(\Psi)\chi(a) \rightarrow \text{Tr}_p \rho_p(\Psi) [V_2, \chi(a)] \quad (4.5)$$

and clearly $B(\Psi)$ depends only on the net polarization of the plasma *inside* the bound electron. It is important to note that the polarization of the plasma in (4.5) is not only due to the monopole interaction ϕ_0 but also to the constraint $\Phi^* = \Psi$. For example, for neutral radiators and weakly coupled plasmas it was shown in Ref. 10 that the ion density is not uniform but instead the ions tend to stay away from the radiator nucleus. Also note that even though the density matrix for the plasma is spherically symmetric about the origin the constrained density matrix $\rho_p(\Psi)$ is not, and therefore, the $l \neq 0$ terms in the definition of V_2 [see Eq. (2.7)] do not vanish.

It must be emphasized that the result in Eq. (4.5) is not unique and depends on the particular choice (4.1) plus the approximation (4.3). Nevertheless, this section clearly

shows the importance of treating plasma correlations self-consistently in line-shift theories. Firstly, it demonstrates the effect of subtracting from the level shifts in $B(\Psi)$ any contributions to the radiator-plasma interaction already included in $L(a; \Psi)$. A comparison of the result in Eq. (4.5) with the equivalent expression for the static shift operator given by the usual theories,¹⁴ which treat electron-ion correlations in *ad hoc* fashion, shows that the two are formally different. The latter involves an electron average over the radiator-electron interaction $v_1^e + v_2^e$ and is independent of the ion configurations. Secondly, by choosing something for Φ^* other than (4.1), for example, let

$$\Phi_k^* \equiv \sum_{j=1}^{N_i} \phi_k^i(j), \quad k=1,2,3,\dots \quad (4.6)$$

so that Φ_k^* is now given by the bare Coulomb ion fields and field derivatives, it is easy to see that the result for $B(\Psi)$ will be different from (4.5). Consequently, different

choices for Φ^* transfer contributions between $L(a; \Psi)$ and $B(\Psi)$.

V. DISCUSSION

The present formalism shows that Stark broadening of spectral lines may be formally expressed as a convolution of static and dynamic broadening. In general, however, a self-consistent treatment of plasma correlations does not allow a clean separation into independent ion and electron broadening so familiar in usual line-shape theories.

There are several new features that are associated with this new formulation. Firstly, all averages occur over a density matrix of the total system so that all plasma interactions can be incorporated in a systematic way. Secondly, the introduction of the ion configuration distribution changes the ensemble for the radiator and plasma from an equilibrium canonical ensemble to the constrained ensemble, Eq. (2.9).

A third feature is the choice of Φ^* . Because the latter is arbitrary, it shows that the level shifts due to time aver-

ages of the radiator-plasma interaction and ion field inhomogeneities are very closely related and that, in fact, some care is necessary in order to avoid overcounting. Although the results in this paper are only formal, they suggest that a self-consistent treatment of plasma correlations requires a much more complete theory than has been used in past calculations of static shifts. In particular, past calculations of level shifts due to the time average of the radiator-plasma interaction must be reconsidered.

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