SPECTRAL-LINE BROADENING AS A PROBLEM OF THE THEORY OF FLUCTUATIONS IN NONEQUILIBRIUM PLASMA

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An expression for the shock profile of a spectral line with allowance for nonelastic processes is obtained within the framework of the kinetic approach. The width and shift operator, corresponding to Stark broadening is governed by the behavior of the spectral density of low-frequency field fluctuations. Due to incorporation of the medium's polarization, the resultant expression does not contain a divergence corresponding to large impact parameters.

The broadening of spectral lines by plasmas is both of experimental and analytic interest [1]. One is usually concerned either with Stark broadening [2] or with broadening due to inelastic transitions [3,4].

In this paper we obtain an expression for the line shape which makes allowance for both these factors and for polarization of the plasma, within the framework of the "kinetic" approach, developed by the first of the present authors [5]. By its spirit this approach is close to the so-called "relaxation" theory [1].

In this approach the line shape is controlled by time dependence of the polarization vector of absorbing atoms, corresponding to transition from degenerated level b to degenerated level a. The polarization vector is expressed in terms of nondiagonal elements of the atomic subsystem density matrix. The expression for the density matrix is found by methods of the kinetic theory of plasma. By virtue of the same fact the kinetic theory of the shape of spectral lines is formulated as one of the directions of the general theory of fluctuations in nonequilibrium plasma. The problem of broadening of laser-emitted spectral lines is solved analogously [6]. As shall be shown below, the line shape is a function of the spectral densities of fluctuations of the longitudinal and transverse fields $(\delta E\,\delta E)^{\parallel}_{\omega,k}$, $(\delta E\,\delta E)^{\perp}_{\omega,k}$, controlled by various processes [7]. This also corrects for their effect on the line shape. The results below are rather general, and their application to specific lines requires numerical methods, which is not considered here.

Statement of the Problem. We shall consider a plasma uniform in space. Let the concentration of charged particles be much greater than the concentration of the atoms: $n_e \gg n_{ei}$. Then broadening due to intrinsic pressure can be neglected. Then, since there are no difficulties in making allowance for Doppler broadening, the atoms can be regarded as immobile without loss of generality.

The polarization vector corresponding to transition $a \to$ b is expressed in terms of density matrix $f_{\alpha\beta}(t)$ as:

$$\mathbf{P}_{ab}(t) = n_{ei} \sum_{\alpha\beta} (\mathbf{d}_{\beta\alpha} f_{\alpha\beta} + \mathbf{d}_{\alpha\beta} f_{\beta\alpha}). \tag{1}$$

Here α and β designate the ensemble of parabolic quantum numbers of levels a and b, respectively.

In order to derive an equation for function $f_{\alpha\beta}(t)$, we shall use as the starting system the equations for the operator density matrix $N_{\nu\mu}(\mathbf{R}, t)$:

$$\left[\frac{\partial}{\partial t} + \frac{i}{\hbar}(\mathcal{H}_{0\nu\nu} - \mathcal{H}_{0\mu\mu})\right] N_{\nu\mu}(\mathbf{R}, t) =$$

$$= \frac{i}{\hbar} \sum_{\nu_1} [\mathbf{d}_{\nu\nu_1} N_{\nu_1\mu}(\mathbf{R}, t) - N_{\nu\nu_1}(\mathbf{R}, t)\, \mathbf{d}_{\nu_1\mu}]\, \mathbf{E}^M(\mathbf{R}, t), \tag{2}$$

$$\mathbf{E}^M = \mathbf{E}^{\perp} + \mathbf{E}_e^{\parallel}.$$

Quantities $\mathcal{H}_{0\nu\nu}$ designate $E_\nu + eFz_{\nu\nu}$, F is the strength of the ion microfield. Summation in Eq. (2) is performed over all the states corresponding to the spectrum of the eigenvalues of the Hamiltonian of an isolated atom.

We designate by symbol $\langle ... \rangle_{hf}$ averaging over the ensemble at a given ionic microfield; whereas the averaging over the distribution of the ionic microfield is designated by $\{...\}_F$.

Be definition,

$$\langle N_{\nu\mu}(\mathbf{R}, t)\rangle_{hf} = n_{ei} f_{\nu\mu}(F, t), \quad f_{\nu\mu}(t) = \{f_{\nu\mu}(F, t)\}_{\mathbf{F}}. \tag{3}$$

Averaging Eqs. (2), we obtain an expression for $f_{\alpha\beta}(F, t)$:

$$\left[\frac{\partial}{\partial t} + \frac{i}{\hbar}(\mathcal{H}_{0\alpha\alpha} - \mathcal{H}_{0\beta\beta})\right] f_{\alpha\beta}(F, t) = I_{\alpha\beta}(F, t), \tag{4}$$

where

$$I_{\alpha\beta}(F, t) = \frac{i}{n_{ei}\hbar} \sum_{\nu} [\mathbf{d}_{\alpha\nu} \langle \delta N_{\nu\mu}\, \delta\mathbf{E}\rangle_{hf} - \langle \delta N_{\alpha\nu}\, \delta\mathbf{E}\rangle_{hf}\, \mathbf{d}_{\nu\mu}]. \tag{5}$$

Equations (1), (3)-(5) can be used for finding the line shape.

The Collision Integral. Dissipation Matrix. The method of derivation of the expression for the collision integral is presented by the first of the present authors [5], and it is not described here. We only note the simplifications used in the calculations.

1. The dependence of the spectral density of the field's fluctuations on the strength of the ionic microfield is not considered.

2. Rapid changes in the collision integral are due to both rapid changes in distribution functions $f_{\mu\nu}$, and by rapid components of the spectral density of the field's fluctuations. The latter do not directly affect the broadening of spectral lines, and hence are not included in the calculations.

3. The expression for the collision integral retains only "resonant" terms, i.e., contributions proportional to $\exp(-i\omega_{ab}t)$, ω_{ab} is the frequency of transition of an isolated atom.

The solution of kinetic equation (4) is written as a Fourier expansion for time functions. We introduce the dissipation matrix, which is represented as the sum $\Phi_{ab}+\Gamma_a+\Gamma^*_b$. Using it we write:

$$I_{ab}(F,\omega)=-[\Phi_{ab}(F,\omega)+\Gamma_a(F)+\Gamma_b^*(F)]f_{ab}(F,\omega). \qquad (6)$$

The right-hand side of this equation is defined as:

$$\Phi_{ab}(F,\omega)=\frac{1}{\hbar^2}\int\frac{d\mathbf{k}}{(2\pi)^3}\int_0^\infty d\tau\, e^{(i\omega-\Delta)\tau}\Big\{(\mathbf{d}_a)_i\exp\Big(-\frac{i}{\hbar}\mathcal{H}_{ab}\tau\Big)[(\mathbf{d}_a)_j - (\mathbf{d}_b^*)_j]-\exp\Big(-\frac{i}{\hbar}\mathcal{H}_{ab}\tau\Big)[(\mathbf{d}_a)_j(\mathbf{d}_b^*)_i-(\mathbf{d}_b^*)_i(\mathbf{d}_b^*)_j]\Big\}(\delta E_j\,\delta E_i)_{\mathbf{k},-\tau}. \qquad (7)$$

Here

$$((\mathbf{d}_a)_i f_{ab})_{\alpha\beta}=\sum_{\alpha'}(\mathbf{d}_{\alpha\alpha'})_i f_{\alpha'\beta},$$

$$((\mathbf{d}_b^*)_j f_{ab})_{\alpha\beta}=\sum_{\beta'} f_{\alpha\beta'}(\mathbf{d}_{\beta'\beta})_j,$$

$$\mathcal{H}_{ab}=\mathcal{H}_{0a}-\mathcal{H}_{0b}.$$

$\mathcal{H}_{0a}$, $\mathcal{H}_{0b}$ have only diagonal matrix elements, equal, respectively, to $\mathcal{H}_{0\alpha\alpha}$, $\mathcal{H}_{0\beta\beta}$. The remaining terms in Eq. (6) have the following matrix elements:

$$[(\Gamma_a+\Gamma_b^*)f_{ab}]_{\alpha\beta}=\sum_{\alpha'}\Gamma_{\alpha\alpha'}f_{\alpha'\beta}+\sum_{\beta'}f_{\alpha\beta'}\Gamma_{\beta'\beta},$$

where

$$\Gamma_{\alpha\alpha'}(F)=\frac{1}{\hbar^2}\sum_{\nu\notin a}\int\frac{d\mathbf{k}\,d\omega}{(2\pi)^4}(\mathbf{d}_{\alpha\nu})_i\times\left[i\left(\omega-\omega_{ab}+\frac{1}{\hbar}\mathcal{H}_{\nu\beta}\right)+\Delta\right]^{-1}(\mathbf{d}_{\nu\alpha'})_j(\delta E_j\,\delta E_i)_{\omega,\mathbf{k}}, \qquad (8)$$

and the expression for $\Gamma_{\beta'\beta}(F)$ is obtained from here by replacing α, α', $\mathcal{H}_{\nu\beta}$ by β', β, and $\mathcal{H}_{\alpha\nu}$, respectively.

The spectral density of fluctuations of the field contained in Eq. (8) is comprised of the longitudinal and transverse parts:

$$(\delta E_j\,\delta E_i)_{\omega,\mathbf{k}}=(\delta E_j\,\delta E_i)^{\perp}_{\omega,\mathbf{k}}+(\delta E_j\,\delta E_i)^{\parallel}_{\omega,\mathbf{k}}.$$

Quantity $(\delta E_j\,\delta E_i)^{\parallel}_{\omega,\mathbf{k}}$ represents the collision relaxation of Stark sublevels, whereas quantity $(\delta E_j\,\delta E_i)^{\perp}_{\omega,\mathbf{k}}$ can be used, for example, to incorporate radiation damping. We note that $\mathrm{Re}\,\Gamma_{\alpha\alpha}$ and $\mathrm{Re}\,\Gamma_{\beta\beta}$ have the meaning of the reciprocals of the lifetimes of components α and β, governed by inelastic processes, whereas $\mathrm{Im}\,\Gamma_{\alpha\alpha}$, $\mathrm{Im}\,\Gamma_{\beta\beta}$ have the meaning of the corresponding shifts in transition frequencies.

The Line Shape with Allowance for Inelastic Processes. On the basis of Eqs. (1), (4) and (6) the line shape is given by the expression:

$$L(\omega)=\mathrm{Sp}\left(\left[\frac{i}{\hbar}\mathcal{H}_{ab}-i\omega+\Phi_{ab}+\Gamma_a+\Gamma_b^*\right]^{-1}\mathbf{d}_{ab}\mathbf{d}_{ba}\right).$$

We rewrite this expression using standard notation [2, p. 468]:

$$L(\omega) = \mathrm{Re}\left\{ \sum_{\alpha\beta\alpha'\beta'} \langle\alpha'|(\mathbf{d}_{ab})_i|\beta'\rangle \langle\beta|(\mathbf{d}_{ba})_i|\alpha\rangle \times \right.$$
$$\left. \times \left\langle\left\langle\alpha\beta\left|\left[\frac{i}{\hbar}(\mathcal{H}_{0a}-\mathcal{H}_{0b}) - i\omega + \Phi_{ab} + \Gamma_a + \Gamma_b^*\right]^{-1}\right|\alpha'\beta'\right\rangle\right\rangle\right\}_F. \quad (9)$$

As examples we shall subsequently consider radiation broadening of spectral lines and broadening by the electron component. The first of these is due to interaction between the atom oscillator with electromagnetic-field oscillators. This interaction can be incorporated by selecting the spectral density of the field's fluctuations in the form [8, page 208]:

$$(\delta E_j\,\delta E_i)^{\perp}_{\omega,\mathbf{k}} = \frac{8\pi^2}{3} c\hbar k\,\delta(\omega - ck)\,\delta_{ij}.$$

Now, using Eq. (8) and neglecting the renormed shift, we obtain

$$\Gamma_{\alpha\alpha'}(F) = \frac{2}{3\hbar c^3} \sum_{\nu\notin a} \mathbf{d}_{\alpha\nu}\mathbf{d}_{\nu\alpha'} \left(\omega_{ab} - \frac{1}{\hbar}\mathcal{H}_{\nu\beta}\right)^3 \Theta\left(\omega_{ab} - \frac{1}{\hbar}\mathcal{H}_{\nu\beta}\right),$$

where $\Theta(x)=1$, if $x \geq 0$ and $\Theta(x)=0$, if $x < 0$. For the Ly = α line this yields the simple result:

$$\Gamma_{\alpha\alpha'} = \frac{2\omega_{ab}^3}{3\hbar c^3}\,\mathbf{d}_{\alpha\beta}\,\mathbf{d}_{\beta\alpha'}.$$

Let us now consider electron-atom collisions. In these calculations we shall neglect the dependence of Φ_{ab}, Γ_a, Γ_b on F. The quantities $\mathcal{H}_{ab}$, $\mathcal{H}_{\nu\beta}$, etc., contained in Eqs. (7)-(9) can be replaced by ω_{ab}, $\omega_{\nu\beta} = (E_\nu - E_\beta)/\hbar$, etc. The spectral density of the field's fluctuations is expressed as [9, page 171]:

$$(\delta E_j\,\delta E_i)_{\omega,\mathbf{k}} = \frac{\delta_{ij}}{3}(\delta \mathbf{E}\,\delta \mathbf{E})_{\omega,\mathbf{k}} = \frac{\delta_{ij}(4\pi)^3 e^2 n_e}{2k^2} \int \frac{\delta(\omega - \mathbf{k}\mathbf{p}/m_e)\,f_e(\mathbf{p})\,d\mathbf{p}}{|\varepsilon(\omega,\mathbf{k})|^2}. \quad (10)$$

It is usually impossible to integrate with respect to k in Eqs. (7)-(8) due to complexity of function $|\varepsilon(\omega,\mathbf{k})|^{-2}$ contained there. We shall hence make some additional assumptions.

1. The momentum distribution of free charged particles is equilibrium distribution.

2. Function $|\varepsilon(\omega,\mathbf{k})|^{-2}$, which incorporates polarization of the medium in the expression $(\delta\mathbf{E}\delta\mathbf{E})_{\omega,\kappa}$, is replaced by a value averaged with respect to ω. This corresponds to introduction, in calculating the correlations, of the effective potential of the interaction between charged particles (see Secs. 47 and 55 in [9]). Under conditions 1 and 2, Eqs. (10) and (8) yields:

$$\Gamma_{\alpha\alpha'} = \frac{(4\pi)^3 e^2 n_e m_e}{6(2\pi m_e \kappa T_e)^{1/2}} \sum_{\nu\notin a} \int \frac{d\mathbf{k}\,d\omega}{(2\pi)^4} \frac{\mathbf{d}_{\alpha\nu}\mathbf{d}_{\nu\alpha'}}{i(\omega - \omega_{\alpha\nu}) + \Delta} \times$$
$$\times k^{-3}[1 + (r_D k)^{-2}]^{-1} \exp\left(-\frac{m_e\omega^2}{2\kappa T_e k^2}\right). \quad (11)$$

The integrand in Eq. (11) can be approximately replaced by 4π/k, but the domain of integration can be limited so that

$$r_{\max}^{-1} < k < r_B^{-1}, \quad r_{\max} = \min\left(r_D, \sqrt{\kappa T_e/m_e\omega_{\alpha\nu}^2}\right),$$

r_W is the Weisskopf radius (is determined from the condition of suitability of the theory of perturbations). Finally, we assume that $\hbar\omega_{\alpha\nu}/\kappa T_e < 1$. As a result, we obtain the following expressions for the real and imaginary parts of $\Gamma_{\alpha\alpha'}$:

$$\mathrm{Re}\,\Gamma_{\alpha\alpha'} = \frac{8\sqrt{2\pi}}{3} \sum_{\nu \notin \alpha} \mathbf{d}_{\alpha\nu}\, \mathbf{d}_{\nu\alpha'}\, \frac{e^2 n_e m_e^{1/2}}{\hbar^2 (\kappa T_e)^{1/2}} \ln \frac{r_{max}}{r_B}, \tag{12}$$

$$\mathrm{Im}\,\Gamma_{\alpha\alpha'} = \frac{1}{\sqrt{\pi}} \mathrm{Re}\,\Gamma_{\alpha\alpha'}.$$

Expressions for $\mathrm{Re}\,\Gamma_{\beta'\beta}$ and $\mathrm{Im}\,\Gamma_{\beta\beta'}$ are analogous.

We now consider Stark broadening. Neglecting the effect of the ionic microfield, we rewrite (7) in the simpler form:

$$\Phi_{ab}(\Delta\omega_{ab}) = \frac{(\mathbf{d}_a - \mathbf{d}_b^*)^2}{3\hbar^2} \int \frac{d\mathbf{k}\, d\omega'}{(2\pi)^4} \frac{(\delta\mathbf{E}\,\delta\mathbf{E})_{\omega',\mathbf{k}}}{i(\omega' - \Delta\omega_{ab}) + \Delta}, \tag{13}$$

$$\Delta\omega_{ab} = \omega - \omega_{ab}.$$

It is seen by comparing this equation with Eq. (8), that, unlike broadening by inelastic transitions, Stark broadening is controlled by the behavior of the spectral density of the field's low-frequency fluctuations. In particular, in the impact region $(\Delta\omega_{ab} = 0)$

$$\Phi_{ab}(0) = \frac{(\mathbf{d}_a - \mathbf{d}_b^*)^2}{6\hbar^2} \int \frac{d\mathbf{k}}{(2\pi^3)} (\delta\mathbf{E}\,\delta\mathbf{E})_{0,\mathbf{k}}.$$

Substituting here Eq. (10), we find, analogously to (12), the width and shift operator:

$$\Phi_{ab}(0) = \frac{8\pi e^2 n_e m_e}{3(2\pi m_e \kappa T_e)^{1/2}} (\mathbf{d}_a - \mathbf{d}_b^*)^2 \ln \frac{r_D}{r_B}, \tag{14}$$

which is identical to the result presented by Lisitsa [2]. As follows from Eqs. (13) and (10), at nonzero mismatch there appears a new cutoff parameter, equal to $v_0/\Delta\omega_{ab}$ $(v_0 = \sqrt{2\kappa T_e/m_e})$, which is no other than the Lewis parameter.

Let us compare the contributions, to the total line width, by different mechanisms, by order of magnitude. The estimates shall be performed by hydrogen. Over the optical frequency range ($\omega_{ab} = 2\cdot10^{15}$ sec^{-1}), at $T_e = 4\cdot10^4$°K and $n_e = 10^{14}$ cm^{-3} we have

$$\mathrm{Re}(\Gamma_{\alpha\alpha'})_{rad} \ll \mathrm{Re}(\Gamma_{\alpha\alpha'})_{inel} \ll \mathrm{Re}\,\Phi_{\alpha\beta},$$

with the ratio of widths being approximately of two orders of magnitude. This means that the mechanism of Stark broadening predominates under the given conditions.

The following should be said in conclusion. The kinetic approach allows calculating the line shape also in nonequilibrium plasma. For this one must use a set of equations for nondiagonal and diagonal elements of the density matrix, with the collision integrals in the equations selected by a rather simple method. This makes it possible to incorporate also the effect of turbulent perturbations on the line shape. The broadening of spectral lines in weakly turbulent plasma was examined within the framework of the relaxation theory by Capes and Voslamber [10].

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