

# Nonlinear interference effects in Stark broadening of ion lines in a dense plasma

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The influence of nonlinear interference effects on the shape of the emission contour  $I(\omega)$  and frequency redistribution function  $R(\omega, \omega')$  is investigated. The analysis is carried out by two methods: 1) by solving the equation set for the density matrix and 2) by applying the kinetic Green's function formalism. In the case of a three-level system both methods yield identical analytic expressions for the functions  $I(\omega)$  and  $R(\omega, \omega')$ . The advantages and shortcomings of the methods are discussed. As a specific example the "forbidden" component of the spectral doublet  $2^1S-4^1P, 2^1S-4^1D$  of helium-like ions is considered. The values of the atomic constants required are calculated using Cowan's program. Qualitative estimates of the contribution of nonlinear interference effects to the "forbidden" and "allowed" components and their ratios for various plasma parameters are presented. The calculations illustrate the importance of taking into account nonlinear interference effects on calculating the emission contour  $I(\omega)$  and rescattering function  $R(\omega, \omega')$  in a hot dense plasma.

## 1. INTRODUCTION

Line radiation of multiply charged ions (MCI) in a hot dense plasma has been intensively studied in recent years<sup>1-5</sup> in connection with problems of obtaining x-ray sources and plasma diagnostics. Of great importance for these problems is the calculation of the MCI spectral line functions which determine the spectral distribution of the radiation and its transport in the medium. The most important of these functions are the emission (absorption) line shape  $I(\omega)$  and the frequency redistribution function  $R(\omega, \omega')$ , which determines the probability of emission of a quantum with frequency  $\omega$  when a quantum is absorbed with frequency  $\omega'$ .

The problem of calculating the functions  $I(\omega)$  and  $R(\omega, \omega')$  in a dense plasma is connected mainly with the need to take account of the Stark broadening of the lines of the radiating ion due to the electric microfields of the surrounding ions and electrons of the plasma. The specifics of the Stark broadening are dictated by the fact that near the radiating level a level polarizing it is always found, which ensures that the magnitude of the Stark splitting is significant. Just such a situation is characteristic both of hydrogen-like ions, where these neighboring levels are separated by the fine structure interval and of more complicated (e.g., helium-like) ions, where the polarization and mixing of states can lead to the appearance of forbidden transitions.<sup>4-7</sup> It is therefore clear that the calculation of the spectral lines should be based on a consistent account of mixing of near-lying states in the ion field of the plasma. This question obviously has a direct analog in the problem of "dynamic" and "static" intensities in the Stark effect (see Ref. 8).

Many of the calculations of the Stark profiles of the ion lines<sup>1-5</sup> have been based, as a rule, on conventional methods of line broadening theory in which only the evolution operators of the radiating states are taken into account, whereas the density matrix, which determines their relative populations, is assumed to be diagonal in some basis. Solutions for two-level systems within the framework of the more consistent approach of Ref. 9, obviously, are also insufficient to describe the mixing processes of interest to us. At the same

time, the effect of taking mixing processes into account in order to obtain a correct description of atomic spectral functions is well-known in laser physics.<sup>10</sup> Here calculations carried out for three-level systems demonstrate the important role of the so-called nonlinear interference effects (NIEF), which are due precisely to the nondiagonal elements of the density matrix of the radiating states, which are not taken into account in the standard theory of Stark broadening.<sup>7</sup>

The aim of the present work is to investigate NIEF for the case of Stark line broadening in plasmas. This study was carried out assuming stationary ions and collisional electrons. This approximation allows one to obtain an analytic solution of the problem for the three-level model consisting of a ground state 1, a radiating state 2, and a polarizing state 3 (Fig. 1).<sup>11</sup> In Fig. 1 the following notation appear:  $A_{21}$ —the probability of the dipole transition  $2 \rightarrow 1$ ;  $A_2(A_3)$ —the total probability of radiative decay from level 2 (3) to the other levels;  $\nu_e$ —the frequency of electron-collisional mixing of levels 2 and 3; and  $\mathbf{d} \cdot \mathbf{F}$ —the interaction with the static ion field.

For the three-level model it is possible to find and to study the contribution of NIEF to the spectral functions  $I(\omega)$  and  $R(\omega, \omega')$  in explicit form. We also note important

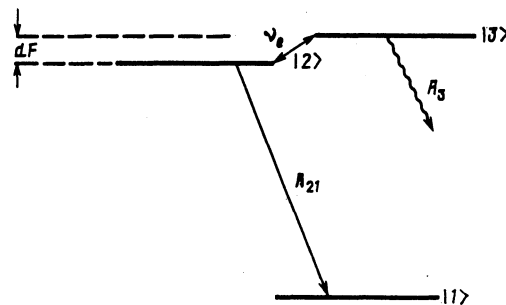


FIG. 1. Level diagram used in the calculations of the emission contour and the rescattering function. The numeration corresponds to the following states of the helium-like ion: 1— $2^1S$ , 2— $4^1P$ , 3— $4^1D$  (concerning the mutual placement of levels 2 and 3 see Sec. 7).

differences between these NIEF's and those encountered in laser physics:<sup>10</sup> 1) the mixing field  $\mathbf{F}$  created by the ions of the plasma is static (in contrast with the resonant field in Ref. 10), 2) in the case of  $R(\omega, \omega')$  there are in reality three fields—the incident field (with frequency  $\omega'$ ), the mixing field ( $\mathbf{F}$ ), and the scattered field (with frequency  $\omega$ ).

The calculations of  $I(\omega)$  and  $R(\omega, \omega')$  are carried out below by two independent methods. The first, widely applied in laser physics,<sup>10</sup> is based on the equations for the atomic density matrix. Its use, however, is associated with a known inconsistency, due to the transition from the compound matrix “atom + electromagnetic field” to the atomic matrix, which requires a nonunique procedure for closing the equation set.<sup>11</sup> Therefore the functions  $I(\omega)$  and  $R(\omega, \omega')$  were also calculated by the more general Green's function method in the Keldysh technique.<sup>12</sup> The results of the two calculations are shown to agree. The way in which we formulate the problem fills a gap in the development of the theory of formation of the line shape and the frequency redistribution function which has been pursued in recent years.<sup>1-15</sup> On the other hand, it is found that in the density matrix method the separation of the contributions to the line shape from the various other physical effects obtains more clearly and requires less cumbersome calculations than in the case of the Green's function technique, wherefore both methods to a certain extent complement each other.

## 2. THE DENSITY MATRIX METHOD

To find the spectral functions  $I(\omega)$  and  $R(\omega, \omega')$  we use the density matrix method.<sup>10</sup> We begin with the general equation in the interaction representation with unperturbed Hamiltonian  $H_0$ :

$$\dot{\rho} = -i(V^{\text{tot}}(t), \rho) + S + R + Q. \quad (2.1)$$

Here  $V^{\text{tot}}(t)$  is the total interaction operator, which is a sum of the interaction with the plasma (ion) field

$$V(t) = [-\exp(-iH_0 t) d\mathbf{F}(t) \exp(iH_0 t)]/\hbar$$

and the interaction with the fields of the incident  $V_1(t)$  and scattered  $V_2(t)$  radiation:

$$V^{\text{tot}}(t) = V(t) + V_1(t) + V_2(t), \quad (2.2)$$

$$V_1(t) = -G_1 \exp(-i\Omega_1 t) \beta_1 \beta_2^+, \quad V_2 = -G_2^* \exp(i\Omega_2 t) \beta_1^+ \beta_2, \quad (2.3)$$

where  $G_1$  and  $G_2$  are the interactions with the field amplitudes,  $\Omega_1 = \omega' - \omega_{21}$  and  $\Omega_2 = \omega - \omega_{21}$  are their frequencies measured from the frequency of the allowed transition  $\omega_{21} = (E_2 - E_1)/\hbar$ , and  $\beta_1$  and  $\beta_2^+$  are the annihilation and creation operators of the atom in the corresponding states. In accordance with the radiation rescattering problem the field  $G_1$  is only absorbed and the field  $G_2$  is only emitted. Therefore the operator  $(V_{1,2}, \rho)$  is defined with Hermitian conjugation:  $(V_{1,2}, \rho) = V_{1,2} \rho - \rho V_{1,2}^+$ .

The reason for the incoherent pumping term  $Q$  in Eqs. (2.1) (e.g., by electronic excitation) is that we intend to use this system to find both the line shape  $I(\omega)$  and the redistribution function  $R(\omega, \omega')$ . The operators  $S$  and  $R$  describe respectively collisional (electronic) and radiative relaxation.<sup>10</sup>

Equation (2.1) with the field interaction Hamiltonian

(2.2), (2.3) in fact determines the density matrix not just of the isolated atom, but the compound “atom + spontaneous electromagnetic field” system in accordance with the choice of selection rules for the perturbations (2.3). The nondiagonal element of such a matrix  $\rho_{21}$  multiplied by  $G_2^*$  determines the power of spontaneous emission of the medium<sup>10</sup>:

$$P(\omega) = -2\hbar\omega \text{Re} \langle iG_2^* \exp(i\Omega_2 t) \rho_{21} \rangle. \quad (2.4)$$

For this reason in what follows we will call the nondiagonal element  $\rho_{21}$  of the density matrix the “spontaneous polarization” of the medium.

The symbol  $\langle \dots \rangle$  in Eq. (2.4) denotes the ensemble average over the radiating ions in the plasma, i.e., over the entire set of stochastic variables entering into the equation for  $\rho_{21}$ , first of all the plasma microfield distribution  $\mathbf{F}$  and the Maxwellian distribution of the velocities of the radiating ions, which determines the Doppler line broadening. Note that the radiated power (2.4) in laser physics is usually defined in the context of constant or monochromatic fields. In this case there can be no doubt that the quantity  $P(\omega)$  is time-independent. However, in the case of a variable plasma microfield, fluctuating in time, the stationarity of  $P(\omega)$  is not obvious beforehand. Another possible way calculating the spectral characteristics of the radiation is to calculate not the power but the total work of the field  $Q_T(\omega)$  after some time interval  $T$ . The work  $Q_T(\omega)$  calculated in this way, being an integral of the power  $P(\omega)$  given by (2.4) over the time interval  $T$  can differ from the quantity obtained by multiplying the stationary power by the interval  $T$ . Nevertheless, for ergodic quasistationary physical systems both of these characteristics should coincide as  $T \rightarrow \infty$ . Indeed, integrating over a sufficiently long time interval  $T \rightarrow \infty$  should give rise to the appearance of a definite oscillation frequency  $\Omega_2$  on the transition  $2 \rightarrow 1$ . This oscillation should also appear when the spontaneous polarization  $\langle \rho_{21}(t) \rangle$  is ensemble-averaged. Therefore the means of calculating the spectral characteristics (in the form of the work or the power) is largely a question of convenience.

The operator  $S$  is expressed through the well-known collisional broadening operator  $\Phi$  in the following way (see Ref. 2; cf. Ref. 7):

$$\begin{aligned} S_{\alpha\alpha'}(\rho) &= \sum_{\alpha_1, \alpha_1'} \exp(i\epsilon t) \langle \alpha\alpha_1^+ | \Phi | \alpha'\alpha_1^+ \rangle \rho_{\alpha_1, \alpha_1'} \\ &= -\frac{4}{3}\pi N_e \frac{e^4}{\hbar^2} \left\langle \frac{1}{v} \right\rangle \Lambda \sum_{\alpha_1, \alpha_1'} \exp(i\epsilon t) \rho_{\alpha_1, \alpha_1'} [(\mathbf{r}\mathbf{r})_{\alpha\alpha_1} \delta_{\alpha'\alpha_1'} \\ &\quad + (\mathbf{r}\mathbf{r})_{\alpha'\alpha_1'} \delta_{\alpha\alpha_1} - 2(\mathbf{r})_{\alpha\alpha_1} (\mathbf{r})_{\alpha'\alpha_1'}], \end{aligned} \quad (2.5)$$

where  $N_e$ ,  $v$ ,  $e$ , and  $m$  are respectively the electron density, the thermal velocity, the charge, and the mass,  $\mathbf{r}$  is the position operator of the atomic electron, and  $\epsilon = \omega_{\alpha\alpha_1} - \omega_{\alpha'\alpha_1'}$ .

The solution of Eqs. (2.1)–(2.3) for spontaneous emission (or rescattering) of the field of frequency  $\Omega_2$  is found by perturbation theory in the parameter  $G_2$ . In the zeroth approximation we have

$$\mathcal{L}\rho^0 + i(V_1, \rho^0) = Q, \quad \mathcal{L}\rho = \partial\rho/\partial t - S - R + i(V(t), \rho). \quad (2.6)$$

For the calculations of the line shape  $I(\omega)$  determined by the pumping  $Q$  the interaction with the incident field  $V_1$  can be neglected, setting

$$\mathcal{L}\rho^{00}=Q. \quad (2.7)$$

On the contrary, to find the function  $R(\omega, \omega')$ , it is important to take the interaction  $V_1$  into account, while the contribution of the pumping term  $Q$  to the excited levels can be neglected. The term containing  $V_1$  can be taken into account using perturbation theory by writing

$$\mathcal{L}\rho^{0i}=-i(V_1, \rho^{00}), \quad \mathcal{L}\rho^{02}=-i(V_1, \rho^{0i}). \quad (2.8)$$

The form of Eqs. (2.8) assumes that the pumping term  $Q$  (i.e., the matrix  $\rho^{00}$ ) determines the population only of the lower (ground) state 1 ( $N_1$ ); the index  $i$  in  $\rho^{0i}$  indicates whether the contribution of  $V_1$  is taken into account to first order (for the polarization) as is usual,<sup>10</sup> or to second order (for the populations). Thus, in the zeroth approximation in  $G_2$  the matrix  $\rho^0$  is equal to the sum  $\rho^0 = \rho^{00} + \rho^{01} + \rho^{02}$  of solutions of Eqs. (2.7)–(2.8), where the individual terms are defined in accordance with what has been said above. Below for the zeroth approximation we will use simply the notation  $\rho^0$  without detailing the individual contributions. The equation for the desired density matrix  $\rho$  determining the emission or rescattering arises in first order in the interaction  $G_2$ :

$$\mathcal{L}\rho^1=-i(V_2, \rho^0(t)). \quad (2.9)$$

For simplicity of notation in the specific calculations below we will write the matrix  $\rho^1$  as well as the original matrix without the index. The operator  $R$  is diagonal in the spherical basis, and its effect on the matrix  $\rho$  was determined in Ref. 10.

The usual way of solving the static line broadening problem is to go over to a new basis of wave functions (the Stark basis), diagonalizing the interaction with the ion field  $F$ . For the case under consideration of MCI line broadening, however, this approach is not always convenient, as a result of the large value of the radiative width  $A_{21}$ , which is comparable with the Stark splitting (see also Sec. 6). Therefore all the matrix elements of the operators (including the pumping  $Q$ ) are given below in the spherical basis.

The solution of system of equations (2.7)–(2.9) for the problems of the line shape  $I(\omega)$  and the redistribution function  $R(\omega, \omega')$  differ markedly in complexity and is worked out separately in Secs. 3 and 4.

### 3. THE SPECTRAL LINE SHAPE

Let us find the spectral line shape  $I(\omega)$  of this three-level system. In the density matrix formalism (Sec. 2) the emitted (absorbed) power  $P(\omega)$  is given by the expression

$$P(\omega)=-2\hbar\omega\text{Re}\langle iG^*\bar{\rho}_{21}\rangle, \quad (3.1)$$

which follows from Eq. (2.4) as a result of the periodic dependence on the frequency of the polarization  $\rho_{21}(t) = \bar{\rho}_{21} \exp(-i(\omega - \omega_{21})t)$ , and the spectral line shape of the transition  $2 \rightarrow 1$ , normalized to unity, has the form

$$I(\omega)=-\frac{1}{\pi\langle\rho_{22}^0\rangle}\frac{\langle\text{Re}(iG^*\bar{\rho}_{21})\rangle}{|G|^2}, \quad (3.2)$$

where  $\rho_{22}^0$  is the stationary population of level 2.

To determine the stationary populations due to collisional pumping at the second ( $Q_2$ ) and third ( $Q_3$ ) levels, and

the polarization  $\rho_{22}^0$  associated with the collisional mixing by the electrons of levels 2 and 3 in the constant electrical field of the ions  $F$ , from Eqs. (2.1)–(2.7) after separating out the oscillating part of  $\rho_{32}^0 = \bar{\rho}_{32}^0 \exp(i\omega_{32}t)$  we arrive at the system of equations

$$\begin{aligned} 2V\text{Im}\bar{\rho}_{32}^0+2\Phi(\rho_{22}^0-\rho_{33}^0)+(A_{21}+\Gamma_{22})\rho_{22}^0-A_{32}\rho_{33}^0=Q_2, \\ -2V\text{Im}\bar{\rho}_{32}^0-2\Phi(\rho_{22}^0-\rho_{33}^0)+(A_{32}+A_{31}+\Gamma_{33})\rho_{33}^0=Q_3, \\ \bar{\rho}_{32}^0(2i\omega_{32}+A)+i8\Phi\text{Im}\bar{\rho}_{32}^0+i2V(\rho_{22}^0-\rho_{33}^0)=0, \end{aligned} \quad (3.3)$$

where  $V = (\mathbf{d} \cdot \mathbf{F})_{23}$  is assumed to be real;  $A = A_{32} + A_{21} + A_{31} + \Gamma_{33} + \Gamma_{22}$ ;  $A_{21}$ ,  $A_{31}$ , and  $A_{32}$  are the radiative widths of the transitions  $2 \rightarrow 1$ ,  $3 \rightarrow 1$ , and  $3 \rightarrow 2$ , respectively;  $\Gamma_{33}$  and  $\Gamma_{22}$  are the homogeneous widths of levels 3 and 2, respectively; and  $2\Phi$  is the matrix element of the electron collisional broadening operator in Eq. (2.5). Eliminating the nondiagonal components of the density matrix  $\rho_{32}$  and  $\rho_{33}$  from Eqs. (3.3), we obtain the kinetic equations for the populations  $\rho_{22}$  and  $\rho_{33}$ , in which in addition to the collisional transitions between the states 2 and 3 also enter transitions under the influence of the Stark field—the so-called field mixing with effective frequency

$$\nu_E = \frac{V^2 A}{\omega_{32}^2 + 2\Phi A + A^2/4}.$$

If the solutions of system (3.3) are known, it is possible to find the polarization  $\bar{\rho}_{21}$  on the basis of Eqs. (2.8)–(2.9) from the system of equations

$$\begin{aligned} -i\Omega\bar{\rho}_{21}+iV\bar{\rho}_{31}+\frac{\Gamma_2}{2}\bar{\rho}_{21}=-G\rho_{22}^0, \\ -i(\Omega-\omega_{32})\bar{\rho}_{31}+iV\bar{\rho}_{21}+\frac{\Gamma_3}{2}\bar{\rho}_{31}=-G\bar{\rho}_{32}^0, \end{aligned} \quad (3.4)$$

where  $\Gamma_2 = A_{21} + \Gamma_{22} + 2\Phi + \Gamma_{11}$ ,  $\Gamma_3 = A_{31} + A_{32} + \Gamma_{33} + 2\Phi + \Gamma_{11}$  and  $\Gamma_{11}$  is the homogeneous line width of the lower level 1.

The polarization  $\bar{\rho}_{21}$ , found from Eqs. (3.4), is equal to

$$\begin{aligned} \bar{\rho}_{21}=-iG\left[\rho_{22}^0+\frac{V\bar{\rho}_{32}^0}{(\omega-\omega_{31})+i\Gamma_3/2}\right] \\ \times\left[\omega-\omega_{21}+\frac{i\Gamma_2}{2}-\frac{V^2}{(\omega-\omega_{31})+i\Gamma_3/2}\right]^{-1}. \end{aligned} \quad (3.5)$$

From Eqs. (3.2)–(3.5) we obtain

$$I(\omega)=\frac{\langle\rho_{22}^0\phi(\omega)\rangle}{\langle\rho_{22}^0\rangle}+I_{\text{non}}(\omega),$$

where  $I_{\text{non}}(\omega)$  is the NIEF contribution to the line shape:

$$\begin{aligned} I_{\text{non}}(\omega)=-\frac{V}{\pi}\frac{1}{\langle\rho_{22}^0\rangle}\text{Im}\left\langle\left(\frac{\bar{\rho}_{32}^0}{\Delta(\omega)}\right)\right\rangle \\ =\left\langle\frac{V^2}{\pi\omega_{32}\Delta_{\text{det}}}(\rho_{22}^0-\rho_{33}^0)\left(\frac{A}{2\omega_{32}}\text{Re}\frac{1}{\Delta(\omega)}+\text{Im}\frac{1}{\Delta(\omega)}\right)\right\rangle \\ \langle\rho_{22}^0\rangle, \end{aligned} \quad (3.6)$$

$$\phi(\omega)=-\frac{1}{\pi}\text{Im}\frac{(\omega-\omega_{31})+i\Gamma_3/2}{\Delta(\omega)}, \quad (3.7)$$

$$\text{Re}\Delta(\omega)=(\omega-\omega_{21})(\omega-\omega_{31})-\frac{\Gamma_3\Gamma_2}{4}-V^2, \quad (3.8)$$

$$\text{Im } \Delta(\omega) = \frac{1}{2} [\Gamma_3(\omega - \omega_{21}) + \Gamma_2(\omega - \omega_{31})], \quad (3.9)$$

$$\Delta_{det} = 1 + \frac{A^2}{4\omega_{32}^2} + \frac{2A\Phi}{\omega_{32}^2}. \quad (3.10)$$

Here  $\rho_{22}^0$  and  $\rho_{33}^0$  are given in explicit form by the expressions

$$\rho_{22}^0 = \frac{Q_2 + Q_3}{A_{21} + \Gamma_{22}} - \frac{A_{31} + \Gamma_{33}}{A_{21} + \Gamma_{22}} \times \frac{Q_3 + (\tilde{V} + 2\Phi)(Q_2 + Q_3)/(A_{21} + \Gamma_{22})}{A_{32} + A_{31} + \Gamma_{33} + (\tilde{V} + 2\Phi)(A_{21} + \Gamma_{33} + \Gamma_{22})/(A_{21} + \Gamma_{22})}, \quad (3.11)$$

$$\rho_{33}^0 = \frac{Q_3 + (\tilde{V} + 2\Phi)(Q_2 + Q_3)/(A_{21} + \Gamma_{22})}{A_{32} + A_{31} + \Gamma_{33} + (\tilde{V} + 2\Phi)(A_{31} + \Gamma_{33} + A_{21} + \Gamma_{22})/(A_{21} + \Gamma_{22})}, \quad (3.12)$$

where  $\tilde{V} = AV^2/(\omega_{32}^2 \Delta_{det})$ .

As can be seen from Eqs. (3.5)–(3.6), the first term, which is proportional to  $\phi$ , describes the spontaneous decay, while the second describes the nonlinear interference effect, which vanishes in the limits  $V \rightarrow 0$  and  $V \rightarrow \infty$  ( $V$  is the average microfield) and is normalized to zero frequency. From the form of the NIEF in Eq. (3.6) it is clear that in the present case it is connected with the imbalance of the populations of levels 3 and 2. It is important to emphasize that the NIEF is proportional to the nondiagonal element of the density matrix  $\bar{\rho}_{32}^0$  (3.5).

From Eq. (3.6) it follows that the line shape depends, generally speaking, on the ratio of the pumps to level 2 and 3 and in this sense it is not a universal characteristic determined only by the evolution of the states of the system. In particular, for pumping by a wideband light source  $Q_3 = 0$  and the NIEF is generally absent. Thus, the line shapes for electronic excitation ( $Q_3 \sim Q_2$ ) and photoexcitation should differ markedly. Mention should be made, however, of the restricted character of the statement of problem (3.3) associated with the very introduction of the pumpings  $Q_2$  and  $Q_3$  as independent parameters. Indeed, the values of  $Q_2$  and  $Q_3$  should be determined by the population kinetics and the mixing of other excited states, so that we arrive at an infinite system of equations. Nevertheless, in a number of cases it is possible to limit oneself to direct pumping to these levels and the given model problem has only limited meaning.

It is easy to go from Eqs. (3.2)–(3.12) to the particular case of the forbidden transition  $3 \rightarrow 1$  (setting  $A_{31} = 0$ ), which traditionally has been of great interest for plasma diagnostics based on the characteristics of the spectrum of forbidden components.<sup>5-7</sup>

The results which have been obtained here can be taken into account in calculations of the intensity distributions of the dielectronic satellites in the spectra of multiply charged ions in a dense plasma.<sup>16</sup> The problem which we have considered models the mixing of populations of the doubly excited levels of the ions due to excitation by the plasma microfield and collisional electronic broadening, taking into account the appearance of additional forbidden components, which has so far not been studied. Thus, it turns out also to be necessary to take NIEF into account in the calculations of the intensity distributions of the dielectronic satellites, used

in the diagnostics of dense plasmas with multiply charged ions.

#### 4. THE RESCATTERING FUNCTION $R(\omega, \omega')$

Calculations of the rescattering function prove to be markedly more complicated than calculations of the line shape. This is due to the appearance of the absorbed radiation frequency  $\omega'$ , which leads to additional oscillations of the polarizations and populations with combination frequencies, primarily with the difference frequency  $\varepsilon = \omega - \omega'$ . Recall that according to Eq. (2.4) the function  $R(\omega, \omega')$  is defined as the power  $P(\omega, \omega')$  of radiation at the frequency  $\omega$  when the transition  $1 \rightarrow 2$  is pumped with frequency  $\omega'$  (and corresponds to a photon energy  $\hbar\omega$  on the transition  $2 \rightarrow 1$ ), normalized by the quantity

$$N = \left\langle \int \int d\omega d\omega' \frac{P(\omega, \omega')}{\hbar\omega} \right\rangle$$

(see Refs. 13 and 14).

Let us write out the systems of equations for  $\rho$  and  $\rho^0$  in the density matrix formalism. Assuming all dependences of the matrix  $\rho^0(t)$  to be periodic ( $\rho_{21}^0 = \bar{\rho}_{21}^0 \exp(i\Omega_1 t)$ ,  $\rho_{32}^0 = \bar{\rho}_{32}^0 \exp(i\omega_{32} t)$ , etc.), for  $A_{31} = A_{32} = 0$  we obtain

$$\begin{aligned} -i\Omega_1 \bar{\rho}_{21}^0 + iV \bar{\rho}_{31}^0 + \Phi \bar{\rho}_{21}^0 + (A_{21} + \Gamma_{22} + \Gamma_{11}) \bar{\rho}_{21}^0 / 2 &= iN_1 G_1, \\ i(\omega_{32} - \Omega_1) \bar{\rho}_{31}^0 + iV \bar{\rho}_{21}^0 + (\Phi + (\Gamma_{33} + \Gamma_{11})/2) \bar{\rho}_{31}^0 &= 0, \\ (A_{21} + \Gamma_{22}) \bar{\rho}_{22}^0 - 2V \text{Im}(\bar{\rho}_{32}^0) + 2\Phi(\rho_{22}^0 - \rho_{33}^0) &= -2\text{Re}(iG_1 \bar{\rho}_{21}^0), \\ i\omega_{32} \bar{\rho}_{32}^0 + iV(\rho_{22}^0 - \rho_{33}^0) + 4\Phi \text{Im} \bar{\rho}_{32}^0 + (A_{21} + \Gamma_{22} + \Gamma_{33}) \bar{\rho}_{32}^0 &= -iG_1 \bar{\rho}_{31}^0, \\ 2V \text{Im}(\bar{\rho}_{32}^0) - 2\Phi(\rho_{22}^0 - \rho_{33}^0) + \Gamma_{33} \rho_{33}^0 &= 0. \end{aligned} \quad (4.1)$$

Equation (2.9) for the main matrix  $\rho$ , after substituting the oscillating solutions for  $\rho^0(t)$  in it, contains on its right-hand side a set of oscillations with various combination frequencies. To reduce it to stationary form we must take the amplitudes of these oscillations into account as additional independent variables, setting (see Refs. 10, and 11)

$$\begin{aligned} \rho_{21}(t) &= r_{21} \exp(-i\Omega_2 t) + \bar{r}_{21} \exp(i(\varepsilon - \Omega_1)t), \\ \rho_{31}(t) &= [r_{31} \exp(-i\Omega_2 t) + \bar{r}_{31} \exp(i(\varepsilon - \Omega_1)t)] \exp(i\omega_{32} t) \quad (4.2) \\ \rho_{11}(t) &= r_1 \exp(-i\varepsilon t) + r_1^* \exp(i\varepsilon t). \end{aligned}$$

Here we put  $\omega = \Omega_2 - \Omega_1$ , the asterisk denotes the complex conjugate, and the bar indicates the additional independent variables.

Using the representation (4.2) to solve Eq. (2.9), we arrive at a system of equations for the elements of the main matrix<sup>2)</sup>

$$\begin{aligned} \left(-i\Omega_2 + \Phi + \frac{A_{21} + \Gamma_{22} + \Gamma_{11}}{2}\right) r_{21} + iV r_{31} - iG_1 r_1 &= -iG_2 \rho_{22}^0, \\ \left(i(\omega_{32} - \Omega_2) + \Phi + \frac{\Gamma_{33} + \Gamma_{11}}{2}\right) r_{31} + iV r_{21} &= -iG_2 \bar{\rho}_{32}^0, \\ (-i\varepsilon + \Gamma_{11}) r_1 &= iG_2 \bar{\rho}_{21}^0. \end{aligned} \quad (4.3)$$

Equations (4.3) describe the complicated interference process of mixing of polarizations and populations as a consequence of the interaction with the external ion field  $V$ . The nature of the evolution of the system is determined by the three elements of the zero-order density matrix  $\rho_{22}^0$ ,  $\bar{\rho}_{32}^0$ , and  $\bar{\rho}_{21}^0$ . To calculate the rescattering spectrum from Eqs. (4.3),

it is necessary to find the parameter  $r_{21}$ , which determines the spontaneous polarization of the medium.

Calculating the number of emitted photons with frequency  $\omega$  and normalizing by  $|G_1|^2|G_2|^2N_1$ , after a number of transformations we obtain

$$R(\omega, \omega') = \frac{1}{I_0} \left\langle \text{Im} \left\{ [q(\omega) + i\phi(\omega)] \times \left[ \tilde{\rho}_{22}^0 - \frac{\tilde{\rho}_{21}^{0*}(-i\Gamma_{11} + (\omega - \omega'))}{(\omega - \omega')^2 + \Gamma_{11}^2} \frac{A}{2} + \frac{\tilde{\rho}_{32}^0 V}{(\omega - \omega_{31}) + i\Gamma_3/2} \right] \right\} \right\rangle. \quad (4.4)$$

In Eq. (4.4) we have introduced the matrix elements  $\tilde{\rho}^0$ , which ensure that Eq. (4.4) is normalized to unity in the  $\omega, \omega'$  plane, and which do not contain  $G_1 N_1$ ;  $A \equiv A_{21} + \Gamma_{22}$ ; and  $I_0$  is a normalizing factor. For  $\Gamma_{33} = 0$  the quantities  $\tilde{\rho}_{ik}^0$  have the form

$$\begin{aligned} \tilde{\rho}_{22}^0 &= \phi(\omega'), \\ \tilde{\rho}_{21}^{0*} &= (q(\omega') - i\phi(\omega')) = \frac{1}{\pi} \frac{i\Gamma_3/2 - (\omega' - \omega_{31})}{\Delta^*(\omega')}, \\ \text{Im } \tilde{\rho}_{32}^0 &= \frac{V}{\Delta_{det}} \frac{A/2\pi}{|\Delta(\omega')|^2} \left( \frac{A}{2} \text{Re } \Delta(\omega') - \omega_{32} \text{Im } \Delta(\omega') \right), \\ \text{Re } \tilde{\rho}_{32}^0 &= \frac{V}{\Delta_{det}} \frac{A/2\pi}{|\Delta(\omega')|^2} \left\{ 2 \left( 2\Phi + \frac{V^2}{2\Phi} \right) + \frac{A}{2} \right\} \\ &\quad \times \text{Im } \Delta(\omega') + \omega_{32} \text{Re } \Delta(\omega'), \end{aligned} \quad (4.5)$$

$$\Delta_{det} = \omega_{32}^2 + A^2/4 + A(2\Phi + V^2/2\Phi),$$

where the functions  $q(\omega)$  and  $\phi(\omega)$  are related by the Kramers-Kronig relation.<sup>17</sup> The physical meaning of the solution (4.4)–(4.5) consists in the following: the term containing  $\tilde{\rho}_{22}^0$  describes the contribution of the spontaneous radiation to the incoherent part of the rescattering function; the term containing  $\tilde{\rho}_{21}^{0*}$  describes the coherent part (in particular, in the limit  $\Gamma_{11} \rightarrow 0$  it contains the unshifted Rayleigh scattering, proportional to  $\delta(\omega - \omega')$ , and also the incoherent part, corresponding to  $P(1/(\omega - \omega'))$ ); and finally, the term containing  $\tilde{\rho}_{32}^0$  describes the nonlinear interference effects (NIEF), taking mixing of states into account.

The next calculation has to do with the substitution of the explicit form of the solution of system (4.1) for the zero-order density matrix. Expressing these solutions in terms of the spectral functions  $\phi(\omega)$ ,  $q(\omega)$ , and  $\Delta(\omega)$ , it is convenient to divide the rescattering function into a “coherent” and an “incoherent” part

$$R(\omega, \omega') = R_{inc}(\omega, \omega') + R_{coh}(\omega, \omega'), \quad (4.6)$$

$$R_{coh} = \frac{1}{\pi} \left\langle [q(\omega)q(\omega') + \phi(\omega)\phi(\omega')] \times \frac{\Gamma_{11}}{(\omega' - \omega)^2 + \Gamma_{11}^2} \frac{A_{21} + \Gamma_{22}}{2} \right\rangle, \quad (4.7)$$

$$R_{inc} = R_{sp} + I_{non}(\omega, \omega') - \left\langle [q(\omega')\phi(\omega) - q(\omega)\phi(\omega')] \times \frac{\omega - \omega'}{(\omega - \omega')^2 + \Gamma_{11}^2} \frac{A_{21} + \Gamma_{22}}{2} \right\rangle, \quad (4.8)$$

In their general form these formulas look quite cumbersome; however, writing the homogeneous width of the upper level (with  $A_{31} = 0$ ) as  $\Gamma_{33} = 0$ , we can obtain the more tractable expressions

$$R_{sp} = \langle \phi(\omega)\phi(\omega') \rangle, \quad (4.9)$$

$$\begin{aligned} I_{non}(\omega, \omega') &= \left\langle -\frac{V}{\pi^2} \text{Im} \left\{ \frac{\tilde{\rho}_{32}^0(\omega')}{\Delta(\omega)} \right\} \right\rangle \\ &= \left\langle -\frac{1}{\pi^2} \frac{V^2}{\Delta_{det}} \frac{1}{|\Delta(\omega)|^2 |\Delta(\omega')|^2} \right. \\ &\quad \times \left\{ \frac{A_{21} + \Gamma_{22}}{2} \text{Re } \Delta(\omega) \text{Re } \Delta(\omega') \right. \\ &\quad \left. \left. - \omega_{32} [\text{Re } \Delta(\omega) \text{Im } \Delta(\omega') + \text{Re } \Delta(\omega') \text{Im } \Delta(\omega)] \right. \right. \\ &\quad \left. \left. - 2 \left( 2\Phi + \frac{V^2}{2\Phi} + \frac{A_{21} + \Gamma_{22}}{4} \right) \text{Im } \Delta(\omega) \text{Im } \Delta(\omega') \right\} \frac{A_{21} + \Gamma_{22}}{2} \right\rangle. \end{aligned} \quad (4.10)$$

In the limit  $\Gamma_{11} \rightarrow 0$  Eq. (4.7) gives the Rayleigh term, and the last term in Eq. (4.8) gives the main contribution according to Ref. 17. Relation (4.9) describes the contribution of the spontaneous emission, while Eq. (4.10) describes the contribution of NIEF to the frequency redistribution function at  $\Gamma_{33} = 0$ . In this formalism the symmetry with respect to the substitution  $\omega \rightarrow \omega'$  is obvious:  $R(\omega, \omega') = R(\omega', \omega)$ .

As is clear from Eqs. (4.6)–(4.10),  $R(\omega, \omega')$  is normalized to unity:

$$\int d\omega d\omega' R(\omega, \omega') = 1. \quad (4.11)$$

The integrals over  $\omega$  and  $\omega'$  in Eq. (4.10) and in the sum in Eq. (4.7) and in the last term in Eq. (4.8) vanish. If we set  $\Gamma_{22} = \Gamma_{11} = 0$ , then in the limit  $\Phi \rightarrow 0$  the only nonzero term is the one corresponding to Eq. (4.7), while the rest of the terms cancel.

Results (4.6)–(4.10) taken together give the explicit analytic form of the rescattering function in the three-level system taking into account relaxation of all three levels ( $\Gamma_{33} = 0$ ), and the collisional transitions between levels 2 and 3 and their field mixing. The final result is obtained after averaging Eq. (4.6) over the distribution function of the ion fields and Doppler shifts  $\omega - \mathbf{k} \cdot \mathbf{v}$  and  $\omega' - \mathbf{k}' \cdot \mathbf{v}'$ .

The reduced form of the solution, obtained by the density matrix method, agrees identically with the result obtained by the Green's-function method (Sec. 5), in spite of their external differences. The general properties of the solution are indicated below (Sec. 5). Here let us pause to compare Eq. (4.6) and the results of Ref. 17. In the static limit coincidence of the general form of both solutions is observed only if NIEF is neglected. In this case, however, the main difference between the two results consists in the fact that in Ref. 17 the functions  $\phi(\omega)$  and  $q(\omega)$ , already averaged over the distribution of fields  $\mathbf{F}$ , figure in, whereas in our solution only the resulting line shape, which is given by the product of these two functions, is subject to averaging. This difference has to do with the use in Ref. 17 of a division of the total average into a product of averages, which is invalid in the statistical case.

The main thrust of the calculation of the rescattering functions in the presence of the Stark effect is the taking into account of NIEF. In essence the presence of significant Stark broadening assumes, as was mentioned earlier, the presence of additional levels near the emitting level. For this reason it is necessary to go beyond the framework of the two-level scheme, and in place of it to allow for the mixing of states, which leads to NIEF.

### 5. THE KINETIC GREEN'S FUNCTION METHOD

To determine the line shape of photon emission under the influence of collisions and the redistribution of frequencies, we can use the diagram technique for nonequilibrium processes developed by Keldysh (Refs. 12 and 18).<sup>3)</sup> The Keldysh technique contains information on the dynamics of the quantum system described by the retarding and accelerating Green's functions  $G^R$  and  $G^A$ , and also on its kinetics, described by the Green's function  $G^{+-}$ . For nondegenerate systems the functions  $G^{+-}$  are expressed in terms of  $G^R$  and  $G^A$  with the help of the relation

$$G^{+-} = G^R - G^A,$$

by virtue of which the equations for the retarding Green's functions form a closed system. The atomic system is described by the set of functions  $G_{ij}^{\alpha\alpha'}$ . For an ideal photon gas the kinetic Green's function  $D_{ij}^{+-}$  has the form<sup>12,18,19</sup>

$$iD_{ij}^{+-}(\omega, \mathbf{k}) = \left( \delta_{ij} - \frac{k_i k_j}{2k^2} \right) (2\pi)^2 \hbar \omega_k [n_k \delta(\omega - \omega_k) + (1 + n_k) \delta(\omega + \omega_k)]. \quad (5.1)$$

Here  $\omega_k = c|\mathbf{k}|$ , and  $n_k$  are the occupation numbers of the photons with respect to momenta, related to the spectral radiation density  $J(\omega_k, \Omega)$  (a function of angles and frequencies) by the relation

$$n_k = \frac{\pi \lambda^2}{\hbar \omega_k} J(\omega_k, \Omega), \quad (5.2)$$

where  $\lambda$  is the wavelength,  $\Omega$  is the unit vector in the direction of propagation of the radiation, thus:  $\Omega = \mathbf{k}/|\mathbf{k}|$ . The radiation intensity  $J(\omega_k, \Omega)$  satisfies the transfer equation, which follows from the Dyson equation for the Green's function  $D_{ij}^{+-}$  (Refs. 12, 18–22) and in the stationary case has the form

$$(\Omega \nabla) J = -k(\omega) J + \varepsilon(\omega). \quad (5.3)$$

The absorption coefficient  $k(\omega)$  and the volume source  $\varepsilon(\omega)$  are expressed in terms of the polarization operators  $\Pi^{+-}$  and  $\Pi^{-+}$  with the help of the atomic Green's functions

$$k(\omega) = -\frac{2\pi i \hbar \omega_k}{c} (\Pi^{+-}(\omega, \mathbf{k}) - \Pi^{-+}(\omega, \mathbf{k})), \quad (5.4)$$

$$\varepsilon(\omega) = -\frac{2i \hbar^2 \omega_k^2}{c \lambda^2} \Pi^{-+}(\omega, \mathbf{k}). \quad (5.5)$$

In the resonance approximation for the polarization operators figuring in Eqs. (5.4)–(5.5), it is possible to obtain

$$\Pi^{+-}(\omega, \mathbf{k}) = i \frac{d_{12}^2}{3} \int \frac{d\omega_p d\mathbf{p}}{(2\pi)^4} G_{22}^{+-}(\omega_p + \omega_k, \mathbf{p} + \mathbf{k}) G_{11}^{+-}(\omega_p, \mathbf{p}), \quad (5.6)$$

$$\Pi^{-+}(\omega, \mathbf{k}) = i \frac{d_{12}^2}{3} \int \frac{d\omega_p d\mathbf{p}}{(2\pi)^4} G_{22}^{-+}(\omega_p + \omega_k, \mathbf{p} + \mathbf{k}) G_{11}^{-+}(\omega_p, \mathbf{p}), \quad (5.7)$$

Thus, in this formalism the spectral photon source is expressed with the help of relations (5.7) in terms of the atomic Green's functions  $G_{22}^{+-}$  and  $G_{11}^{+-}$ . The atomic density matrix is expressed in terms of the frequency-integrated functions  $G_{ik}^{+-}(\mathbf{p}, \omega)$ :

$$\rho_{ik}(\mathbf{p}) = -i \int G_{ik}^{+-}(\mathbf{p}, \omega) \frac{d\omega}{2\pi}. \quad (5.8)$$

To find the Green's functions  $G_{ik}^{\alpha\alpha'}$  we make use of the resonance approximation in the Dyson equations. We write the Dyson equation for the retarding Green's functions  $G_{ik}^R$  in the static Stark field  $\mathbf{F}$ , assuming that  $G_{ik}^R$  depend only on  $\omega$  and  $\mathbf{F}$ :

$$\begin{aligned} (\omega - \omega_2 - \varepsilon_p) G_{22}^R &= 1 - V G_{32}^R + \Sigma_{22}^R G_{22}^R + \Sigma_{23}^R G_{32}^R, \\ (\omega - \omega_3 - \varepsilon_p) G_{32}^R &= -V G_{22}^R + \Sigma_{33}^R G_{32}^R + \Sigma_{32}^R G_{22}^R. \end{aligned} \quad (5.9)$$

It is possible to write analogous equations for the functions  $G_{33}^R$  and  $G_{23}^R$ .

Assuming the level shifts to be included in the corresponding frequencies  $\omega_i$ , we obtain for the values of the mass operators figuring in Eq. (5.9)

$$\Sigma_{22}^R = -i\Gamma_2/2, \quad \Sigma_{33}^R = -i\Gamma_3/2, \quad \Sigma_{23}^R = \Sigma_{32}^R = 0,$$

where  $\Sigma_{ik}^R = -(1/2)\Sigma_{ik}^{+-}$ , and the  $\Sigma_{ik}^{+-}$  are represented in the form shown in Figs. 2a and b.<sup>19</sup>

The first diagram describes the radiative decay  $2 \rightarrow 1$ ; the second, collisional mixing of the states  $3 \leftrightarrow 2$  under the action of the electrons. The wavy lines represent the photon Green's functions, the thick line, the atomic Green's functions, the dashed line, the interaction potential of the atomic electrons with the broadening particles, whose Green's function is denoted by the thin solid line. Using the expression for  $D^{+-}(q)$  (5.1), we obtain

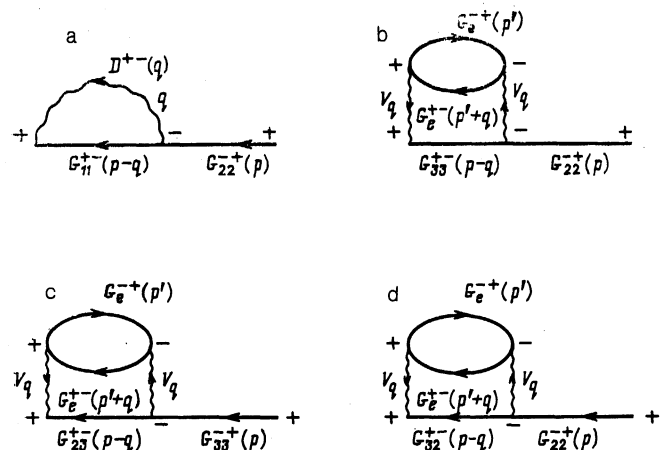


FIG. 2. Diagram representation of the mass operators: a)  $\Sigma_{22}^{+-}$ , interaction with the photon field taken into account, b)  $\Sigma_{33}^{+-}$ , collisional interactions taken into account, c)  $\Sigma_{23}^{+-}$ , collisional interactions taken into account, d)  $\Sigma_{32}^{+-}$ , collisional interactions taken into account.

$$\Gamma_2 = A + \nu_{32}, \quad (5.10)$$

$$\Gamma_3 = \nu_{32},$$

where  $A$  is the probability of spontaneous emission on the transition  $2 \rightarrow 1$ , and  $\nu_{32}$  is the frequency of collisional mixing ( $\nu_{32} = \nu_{23} = \nu$ ):

$$\nu = 2\pi \int \frac{d\mathbf{p}' d\mathbf{q}}{(2\pi)^6} n_{\mathbf{p}'} \delta(\omega_{\mathbf{p}} - \varepsilon_{\mathbf{p}'} - \varepsilon_{\mathbf{q}} - \mathbf{q}\mathbf{v}' - \omega_{32}) |V_{\mathbf{q}}|^2. \quad (5.11)$$

Here  $n_{\mathbf{p}}$  are the occupation numbers of the electrons in momentum space. The electron temperature is assumed to be large in comparison with  $\omega_{32}$  and  $V$ . The nondiagonal elements  $\Sigma_{23}$  and  $\Sigma_{32}$  are proportional to the diagrams shown in Figs. 2c and d, and are characterized by an integral of the form

$$\int G_{32}^{+-}(\mathbf{p}-\mathbf{q}) d\omega_{\mathbf{q}},$$

which can be represented in the form

$$\int (G_{32}^R - G_{32}^A) d\omega_{\mathbf{q}} = 0. \quad (5.12)$$

Relation (5.12) follows from the expressions for  $G_{32}^{R,A}$  derived below and the convergence of the integrals in each of the terms in Eq. (5.12). From Eqs. (5.9)–(5.12) we obtain

$$G_{22}^R(\omega, \mathbf{p}) = \frac{\omega - \omega_{31} - \varepsilon_{\mathbf{p}} + i\Gamma_3/2}{(\omega - \omega_{21} - \varepsilon_{\mathbf{p}} + i\Gamma_2/2)(\omega - \omega_{31} - \varepsilon_{\mathbf{p}} + i\Gamma_3/2) - V^2}, \quad (5.13)$$

$$G_{32}^R(\omega, \mathbf{p}) = -\frac{VG_{22}^R(\omega, \mathbf{p})}{\omega - \omega_{31} - \varepsilon_{\mathbf{p}} + i\Gamma_3/2}, \quad (5.14)$$

$$G_{ik}^A(\omega) = (G_{ik}^R(\omega))^*.$$

In what follows for simplicity we will omit the translational energy of the atomic particles  $\varepsilon_{\mathbf{p}}$ . For  $G_{33}^R(\omega)$  analogously we obtain

$$G_{33}^R(\omega) = \frac{\omega - \omega_{21} + i\Gamma_2/2}{\Delta(\omega)}, \quad (5.15)$$

$$\Delta(\omega) = (\omega - \omega_{21} + i\Gamma_2/2)(\omega - \omega_{31} + i\Gamma_3/2) - V^2.$$

The elementary line shape  $a_{22}(\omega)$ ,  $a_{33}(\omega)$  is given by the expressions

$$\begin{aligned} G_{ii}^R - G_{ii}^A &= -2\pi i a_{ii}(\omega), \\ a_{22}(\omega) &= -(1/\pi) \text{Im} \frac{\omega - \omega_{31} + i\Gamma_3/2}{\Delta(\omega)}, \\ a_{33}(\omega) &= -(1/\pi) \text{Im} \frac{\omega - \omega_{21} + i\Gamma_2/2}{\Delta(\omega)}. \end{aligned} \quad (5.16)$$

Assuming as before that the matrix element of the interaction with the "Stark" field  $V$  is real, we have  $G_{23}^R(\omega) = G_{32}^R(\omega)$ .

For the kinetic Green's functions  $G_{jk}^{-+}(\omega)$  from the Dyson equations (cf. Refs. 12 and 18) we find the following system:

$$\begin{aligned} \left( \frac{\partial}{\partial T} - \omega_j + \omega_k \right) G_{jk}^{-+} \\ = -V_{js} G_{sk}^{-+} + G_{js}^{-+} V_{sk} + \Sigma_{js}^{-\alpha} G_{sk}^{\alpha+} + G_{js}^{-\alpha} \Sigma_{sk}^{\alpha+}, \end{aligned} \quad (5.17)$$

The indices  $s$  (2,3) and  $\alpha$  ( $\pm$ ) are understood to be summed over.

Using relation (5.8), departing from Eq. (5.17) and taking into account relations (5.16) we write out the system of equations<sup>4)</sup>

$$\begin{aligned} i \frac{\partial G_{33}^{-+}}{\partial T} + i\nu G_{33}^{-+} &= -(\nu\rho_{22} + Q_3) 2\pi a_{33} - V(G_{23}^{-+} - G_{32}^{-+}) \\ &\quad + i\nu(\rho_{23} G_{23}^A - \rho_{32} G_{32}^R), \\ i \frac{\partial G_{22}^{-+}}{\partial T} + i(\nu + A) G_{22}^{-+} \\ &= -(\nu\rho_{33} + Q_2) 2\pi a_{22} - V(G_{32}^{-+} - G_{23}^{-+}) \\ &\quad + i\nu(\rho_{32} G_{32}^A - \rho_{23} G_{23}^R), \\ \left( i \frac{\partial}{\partial T} - \omega_{32} + i(\nu + A/2) \right) G_{32}^{-+} \\ &= -V(G_{22}^{-+} - G_{33}^{-+}) + i(\rho_{22}\nu + Q_3) G_{32}^A \\ &\quad - i(\rho_{33}\nu + Q_2) G_{32}^R + i\nu\rho_{32}(G_{22}^A - G_{33}^R), \\ \left( i \frac{\partial}{\partial T} + \omega_{32} + i(\nu + A/2) \right) G_{23}^{-+} \\ &= V(G_{22}^{-+} - G_{33}^{-+}) + i(\nu\rho_{33} + Q_2) G_{23}^A \\ &\quad - i(\rho_{22}\nu + Q_3) G_{23}^R - i(\rho_{22}\nu + Q_3) G_{23}^R + i\nu\rho_{32}(G_{33}^A - G_{22}^R). \end{aligned} \quad (5.18)$$

Here  $Q_{2,3}$  are the rates of formation of the atomic particles on levels 2 and 3. It is assumed that the transition  $1 \rightarrow 3$  is dipole-forbidden, for which reason in  $Q_3$ , in contrast with  $Q_2$ , photoabsorption from the nearest lower level does not contribute:  $Q_2 = Q_{2,s} + Q_2(\omega)$ , where  $Q_{2,s}$  is the collisional pumping of the second level,

$$\begin{aligned} Q_2(\omega) &= (\lambda^2/4) AN_1 (g_2/g_1) \\ &\quad \times \int d\Omega \int d\omega_k J(\omega_k, \Omega) f_M(\mathbf{v}) \delta(\omega - \omega_k + \mathbf{k}\mathbf{v}). \end{aligned} \quad (5.19)$$

Here  $\lambda$  is the wavelength of the resonant photon,  $N_1$  is the density of particles in the ground state,  $g_i$  is the static weight of the state  $i$ , and  $f_M(\mathbf{v})$  is the Maxwellian velocity distribution function of the atoms in the ground state over. Taking Eq. (5.8) into account and integrating Eqs. (5.18) over frequency, we obtain

$$\begin{aligned} \left( i \frac{\partial}{\partial T} - \omega_{32} + i \frac{\Gamma_{32}}{2} \right) \rho_{32} &= -V(\rho_{22} - \rho_{33}) + i\nu\rho_{23} - Q, \\ \left( i \frac{\partial}{\partial T} + \omega_{32} + i \frac{\Gamma_{32}}{2} \right) \rho_{23} &= V(\rho_{22} - \rho_{33}) + i\nu\rho_{32} + Q^*, \\ \left( i \frac{\partial}{\partial T} + i\nu \right) \rho_{33} &= i\nu\rho_{22} + iQ_3 - V(\rho_{23} - \rho_{32}), \\ \left( i \frac{\partial}{\partial T} + i(\nu + A) \right) \rho_{22} &= i\nu\rho_{33} + i\bar{Q}_2 + V(\rho_{23} - \rho_{32}), \end{aligned} \quad (5.20)$$

where  $\Gamma_{23} = \Gamma_3 + \Gamma_2$  and

$$Q = \int \frac{d\omega}{2\pi} Q_2(\omega) G_{32}^R(\omega) = -V \int \frac{d\omega}{2\pi} \frac{Q_2(\omega)}{\Delta(\omega)} = -V\kappa, \quad (5.21)$$

$$\bar{Q}_2 = Q_{2,s} + \int d\omega Q_2(\omega) a_{22}(\omega). \quad (5.22)$$

In the case of collisional pumping ( $Q_3$ ,  $Q_{2,s}$ ) only, from

Eqs. (5.18) taking into account Eqs. (5.20) at  $Q = 0$  and  $\bar{Q}_2 = Q_{2,s}$  we have the stationary solution:

$$G^{-+}(\omega) = i \left( 2\pi a_{22}(\omega) \rho_{22}^* + 2V \operatorname{Im} \frac{\rho_{32}^*}{\Delta(\omega)} \right), \quad (5.23)$$

where  $\rho_{ik}^s$  is essentially the solution of system (5.20) at  $Q = 0$  and  $\bar{Q}_2 = Q_{2,s}$ . Solutions of system (5.20)  $\rho_{ik}^s$  completely coincide with the analogous solutions for the atomic density matrix obtained from system (3.3) [see Eqs. (3.11) and (3.12)].

Substituting Eq. (5.23) in Eqs. (5.16) and making use of the relation

$$G_{11}^{+-}(\omega, \bar{\mathbf{p}}) = -2\pi i \delta(\omega_p - E_p + \mu), \quad (5.24)$$

where  $\mu$  is the chemical potential of the atoms, we arrive at an expression for the emission line contour (3.6)–(3.12) due to collisional excitation at  $A_{31} = \Gamma_{33} = A_{32} = \Gamma_{11} = 0$ .

To calculate the rescattering function we seek the stationary solution of Eqs. (5.18) and (5.20) at  $Q_3 = G_{2,s} = 0$ . Then

$$\begin{aligned} G_{22}^{-+}(\omega) = i \left\{ 2\pi a_{22}(\omega) \rho_{22}^\phi + \frac{1}{\Delta_{det}} \left[ \frac{\Gamma_3}{\Gamma_{32}} \bar{v}_E (2\omega_{32} \operatorname{Im}(1/\Delta(\omega)) \right. \right. \\ \left. \left. + \Gamma_{32} \operatorname{Re}(1/\Delta(\omega)) \right) + (v + \bar{v}_E) 2\pi a_{22}(\omega) \right] (Q_2(\omega) - \bar{Q}_2) \\ - \frac{1}{\Delta_{det}} \left[ 2\pi a_{22}(\omega) \frac{2\Gamma_3 V^2}{\omega_{32}^2 + (\Gamma_{32}/2)^2} (\omega_{32} \operatorname{Im}(\kappa) + (\Gamma_{32}/2) \operatorname{Re}(\kappa)) \right. \\ \left. - \frac{\Gamma_3 V}{\omega_{32}^2 + (\Gamma_{32}/2)^2} \left( 4 \operatorname{Im} \rho_{32}^\phi \left[ \operatorname{Im}(\omega_{32}/\Delta(\omega)) + \operatorname{Re} \frac{\Gamma_{32}}{2\Delta(\omega)} \right] \right. \right. \\ \left. \left. \times \left( V^2 - \frac{\Gamma_2 \Gamma_3}{2} \right) + \operatorname{Re} \rho_{32}^\phi \left[ (\Gamma_3 \Gamma_2 + 4V^2) \Gamma_{32} \right. \right. \right. \\ \left. \left. \left. - 2\Gamma_2 \Gamma_3 \omega_{32} \operatorname{Re} \frac{1}{\Delta(\omega)} \right] \right) \right] \right\}. \quad (5.25) \end{aligned}$$

The elements  $\rho_{ik}^\phi$  in Eq. (5.22) have the form

$$\begin{aligned} \rho_{22}^\phi = \bar{Q}_2/A, \\ \operatorname{Im} \rho_{32}^\phi = -\frac{\Gamma_3 V}{D} \left( \omega_{32} \operatorname{Im} \kappa + \frac{A}{2} \operatorname{Re} \kappa \right), \\ \operatorname{Re} \rho_{32}^\phi = -\frac{\omega_{32} V}{D} \left[ -\frac{2V^2}{D} \left( \omega_{32} \operatorname{Im} \kappa + \frac{A}{2} \operatorname{Re} \kappa \right) \right] \\ - \frac{V}{D} \left( \omega_{32} \operatorname{Re} \kappa - \frac{A}{2} \operatorname{Im} \kappa \right) + \frac{2\Gamma_3 V}{D} \operatorname{Im} \kappa. \quad (5.26) \end{aligned}$$

In Eqs. (5.25) and (5.26) we have introduced the notation

$$\Delta_{det} = \Gamma_{32} \bar{v}_E + \Gamma_2 \Gamma_3, \quad (5.27)$$

$$\bar{v}_E = \Gamma_{32} V^2 / (\omega_{32}^2 + (\Gamma_{32}/2)^2), \quad (5.28)$$

$$D = \omega_{32}^2 + A^2/4 + v_A, \quad (5.29)$$

$$\Gamma = \Gamma_3 + v_E, \quad (5.30)$$

$$v_E = V^2 A/D. \quad (5.31)$$

Introducing for definiteness the frequency redistribution function  $R(\omega, \omega')$  from Ref. 13, from Eqs. (5.5) and (5.7) we obtain

$$\epsilon(\omega) = \frac{A\lambda^2}{4} N_1 \frac{g_2}{g_1} \int \frac{d\omega' d\Omega J(\omega', \Omega)}{4\pi\hbar\omega'} R(\omega', \omega), \quad (5.32)$$

where  $R(\omega, \omega') = R(\omega', \omega)$  and

$$R(\omega, \omega') = \int d\mathbf{v} f_M(\mathbf{v}) \langle Q(\omega - \mathbf{k}\mathbf{v}, \omega' - \mathbf{k}\mathbf{v}') \rangle, \quad (5.33)$$

( $\langle \dots \rangle$  denotes averaging over the microfields), and, comparing Eq. (5.32) with Eqs. (5.5) and (5.7), taking Eqs. (5.25)–(5.30) into account, it is possible to find  $Q(\omega, \omega')$ , which can be represented in the form<sup>5)</sup>

$$Q(\omega, \omega') = Q_{inc}(\omega, \omega') + Q_{coh} \delta(\omega - \omega'). \quad (5.34)$$

The first term characterizes the contribution of the incoherent scattering, the second, the coherent. The explicit expression for the coherent part has the form

$$Q_{coh}(\omega) = \frac{A}{\Gamma_2} \left( \phi(\omega) - \frac{\Gamma_3 V^2}{2\pi |\Delta(\omega)|^2} \right), \quad (5.35)$$

where

$$\phi(\omega) = a_{22}(\omega) = \frac{1}{2\pi} \frac{\tilde{\phi}(\Omega)}{|\Delta(\omega)|^2},$$

$$\tilde{\phi}(\Omega) = \Gamma_2 (\Omega - \omega_{32})^2 + \Gamma_3 \left( V^2 + \frac{\Gamma_2 \Gamma_3}{4} \right),$$

$$\Omega = \omega - \omega_{21} = \omega - \omega_2.$$

For the incoherent part  $Q(\omega, \omega')$  we have

$$\begin{aligned} Q_{inc}(\omega, \omega') = \frac{\Gamma_3}{\Gamma_2} \frac{(D + AV^2/\Gamma_2)}{(D + AV^2/\Gamma_3)} \phi(\omega) \phi(\omega') \\ + \frac{A\Gamma_3 V^2}{4\pi^2 \Gamma_2^2} \frac{1}{\Gamma D} \frac{P(\omega, \omega')}{|\Delta(\omega)|^2 |\Delta(\omega')|^2}, \quad (5.36) \end{aligned}$$

where

$$\begin{aligned} P(\omega, \omega') = \left( \Gamma_3 \Gamma_2 \omega_{32}^2 + \Gamma_{32} A \left( V^2 + \frac{\Gamma_2 \Gamma_3}{4} \right) \right) \\ \times \left\{ \Gamma_2 [(\Omega - \omega_{32})^2 + (\Omega' - \omega_{32})^2 + \Gamma_3^2] \right. \\ \left. + \Gamma_3 V^2 \right\} + \omega_{32} \Gamma_3 \left( \tilde{\phi}(\Omega) \frac{\partial \tilde{\phi}(\Omega')}{\partial \Omega'} + \tilde{\phi}(\Omega') \frac{\partial \tilde{\phi}(\Omega)}{\partial \Omega} \right) \\ + \frac{1}{2} (\Gamma_2 \omega_{32}^2 + \Gamma_{32} (V^2 + \Gamma_3 (A/4 + \Gamma_3))) \frac{\partial \tilde{\phi}(\Omega)}{\partial \Omega} \frac{\partial \tilde{\phi}(\Omega')}{\partial \Omega'} \\ + \Gamma_3^2 (V^2 - \Gamma_3 \Gamma_2/2) \omega_{32} \left( \frac{\partial \tilde{\phi}(\Omega)}{\partial \Omega} + \frac{\partial \tilde{\phi}(\Omega')}{\partial \Omega'} \right). \quad (5.37) \end{aligned}$$

Expressions (5.34)–(5.37) coincide with the results of the calculation of the density matrix of the compound system (4.6)–(4.11) in the case  $\Gamma_{11} = \Gamma_{22} = A_{32} = 0$  with the substitution  $2\Phi = v$ ,  $A = A_{21}$ . Here

$$\int Q(\omega, \omega') d\omega' = \phi(\omega). \quad (5.38)$$

If further we have  $\Gamma_3 = 0$ , i.e., mixing of states 2 and 3 due to collisions with the electrons is absent, then

$$Q(\omega, \omega') = \phi(\omega) \delta(\omega - \omega'). \quad (5.39)$$

In the limit  $V \rightarrow 0$  from Eqs. (5.34)–(5.37) we obtain<sup>13)</sup>

$$Q(\omega, \omega') = \frac{v}{v+A} \phi_0(\omega) \phi_0(\omega') + \frac{A}{v+A} \delta(\omega - \omega') \phi_0(\omega). \quad (5.40)$$

Here



$$\phi_0(\omega) = \frac{\Gamma_2}{2\pi(\Omega^2 + (\Gamma_2/2)^2)}.$$

For  $V \ll \Gamma_2, \Gamma_3, \omega_{32}$  and  $\Omega, \Omega' \gg V$  we obtain from Eq. (5.36)

$$Q_{inc}(\omega, \omega') = \Gamma_3^2 / (4\pi^2 \Omega^2 \Omega'^2). \quad (5.41)$$

$$Q_{inc}(\omega, \omega') = \frac{v^2}{4\pi^2} \frac{(\Omega^2 + V^2)(\Omega'^2 + V^2) + 4V^2\Omega\Omega' + \frac{A}{v}V^2(\Omega + \Omega')^2 + O(V^2v^2)}{|\Omega^2 - V^2 + i\Omega v| |\Omega'^2 - V^2 + i\Omega' v|^2}. \quad (5.42)$$

As follows from this expression, the rescattering function has two peaks in both frequencies:  $\Omega = \pm V, \Omega' = \pm V$ . Note that according to Eq. (5.42) the contributions to the rescattering from the peaks corresponding to the condition  $\Omega\Omega' = -V^2$  are suppressed by a factor of more than  $(A/v)^{-1} \gg 1$  in comparison with the contributions from the peaks for which  $\Omega\Omega' = V^2$ . This suppression is a consequence of NIEF, without an account of which<sup>9</sup> the contributions from both peaks to the function  $Q_{inc}(\omega, \omega')$  in a strong field are equal, and the function  $Q_{inc}(\omega, \omega')$  itself is proportional to the product of the line shapes  $\phi(\omega)\phi(\omega')$ .

In weak fields for  $\omega_{32} \gg V$  and  $V \gg A, V \gg v, A \gg v$  it is possible to obtain an approximate expression for  $Q_{inc}(\omega, \omega')$  in the vicinity of the peak  $\Omega = \Omega' = \omega_{32}$ :

$$Q_{inc}(\omega_{32}, \omega_{32}) = \frac{1}{4\pi^2} \frac{A v V^4}{|\Delta|^4} \frac{\omega_{32}^2}{\omega_{32}^2 + A V^2 / v} \left\{ \frac{v^2}{A^2} + \frac{v}{A} + \frac{V^2}{\omega_{32}^2} \right\} \quad (5.43)$$

(here  $|\Delta|^2 = \omega_{32}^2 v^2 / 4 + V^4$ ). It can be easily seen that the second two terms in braces on the right-hand side of Eq. (5.43) represent the NIEF contribution to the frequency redistribution function and substantially exceed the first term. Thus, the influence of NIEF on the height of the peak at  $\Omega = \Omega' = \omega_{32}$  is substantial at small plasma densities.

Neglecting the NIEF contribution we can write an explicit expression for the incoherent rescattering function, analogous to Eqs. (5.36) and (5.37). From Eqs. (4.4), (4.5), (4.7), and (4.8) we obtain

$$\tilde{Q}_{inc}(\omega, \omega') = \frac{\tilde{P}(\omega, \omega')}{4\pi^2 |\Delta(\omega)|^2 |\Delta(\omega')|^2}, \quad (5.44)$$

$$\begin{aligned} \tilde{P}(\omega, \omega') &= \Gamma_3 \Gamma_2 [(\Omega - \omega_{32})^2 + (\Gamma_3/2)^2 + V^2] [(\Omega' - \omega_{32})^2 + (\Gamma_3/2)^2 + V^2] \\ &\quad - A V^2 \{ \Gamma_3 [\Omega'(\Omega' - \omega_{32}) + \Omega(\Omega - \omega_{32})] \\ &\quad + \Gamma_{32}(\Omega - \omega_{32})(\Omega' - \omega_{32}) \} + (A V \Gamma_3/2)^2. \end{aligned} \quad (5.45)$$

From Eqs. (5.44) and (5.45) it follows that in the vicinity of the points  $\Omega' \approx \Omega \approx \omega_{32}$  (but not at the point  $\Omega' = \Omega = \omega_{32}$  itself) the function  $\tilde{P}(\omega, \omega')$  can take on negative values. This is possible if the conditions  $\omega_{32} \gg V \gg (A, v)$ ,  $A \gg v$ , and  $V^2 \gg \omega_{32}^2 v / A$  hold. For  $V \gg \omega_{32}$  the function  $\tilde{P}$  is positive.

If we take NIEF into account the total redistribution function  $Q_{inc}(\omega, \omega')$  is positive. This once again emphasizes that ignoring NIEF can lead to physically meaningless results (cf. Refs. 14 and 17).

The expressions for the spectra of emission with incoherent pumping and the rescattering function found in this

Let us consider the limit of high plasma densities when  $\omega_{32}$  can be neglected in comparison with  $V$  and  $v$ , and  $V \gg v \gg A$  holds. In this case for the function  $Q_{inc}(\omega, \omega')$  we obtain

section reflect the contributions of nonlinear interference effects, which are represented by the second term in Eq. (5.23) and the group of terms containing  $\kappa$  and  $\rho_{32}^{\phi}$  in Eq. (5.25).

The Green's function method used in the present section, in comparison with the method of the density matrix of the compound system, is suitable when the problems of determining the line shape and the frequency redistribution function reduce to the solution of the same system of equations. At the same time, in the density matrix method the actual structure of the final result turns out to be simpler for distinguishing effects of different nature.

## 6. NIEF IN SPHERICAL AND PARABOLIC BASES: ANALYTIC ESTIMATE OF THE CONTRIBUTION TO THE LINE SHAPE

In the theory of Stark broadening it is customary to use the parabolic basis which diagonalizes the atom-electric field interaction.<sup>7</sup> It is related to the spherical basis by a unitary transformation  $\hat{U}$  which for the three-level scheme has the form

$$\hat{U} = \begin{bmatrix} 1 & 0 & 0 \\ 0 & \cos \beta/2 & \sin \beta/2 \\ 0 & -\sin \beta/2 & \cos \beta/2 \end{bmatrix}, \quad (6.1)$$

where  $\beta \equiv \arctan(2V/\omega_{32})$ . In this basis of such Stark compound states the equations for the density matrix (3.3) and (3.4) take the form

$$\begin{aligned} -2A' \operatorname{Re} \tilde{\rho}_{32} + \tilde{v} [\tilde{\rho}_{22} - \tilde{\rho}_{33} - 2(\operatorname{Re} \tilde{\rho}_{32}) \operatorname{tg} \beta] + \tilde{A}_2 \tilde{\rho}_{22} &= \tilde{Q}_2, \\ -2A' \operatorname{Re} \tilde{\rho}_{32} - \tilde{v} [\tilde{\rho}_{22} - \tilde{\rho}_{33} - 2(\operatorname{Re} \tilde{\rho}_{32}) \operatorname{tg} \beta] + \tilde{A}_3 \tilde{\rho}_{33} &= \tilde{Q}_3, \\ (2i\tilde{\omega}_{32} + \tilde{A}_2 + \tilde{A}_3 + 4v) \tilde{\rho}_{32} + 2\tilde{v} [(\tilde{\rho}_{33} - \tilde{\rho}_{22}) \operatorname{tg} \beta - 2 \operatorname{Re} \tilde{\rho}_{32}] \\ - 2A' (\tilde{\rho}_{22} + \tilde{\rho}_{33}) &= (\tilde{Q}_3 - \tilde{Q}_2) \operatorname{tg} \beta, \\ \left[ -i(\omega - \tilde{\omega}_{21}) + \frac{\Gamma_{11} + \tilde{A}_2 + v}{2} \right] \tilde{\rho}_{21} - A' \tilde{\rho}_{31} &= -G_2 \tilde{\rho}_{22} - G_3 \tilde{\rho}_{23}, \\ \left[ -i(\omega - \tilde{\omega}_{31}) + \frac{\Gamma_{11} + \tilde{A}_3 + v}{2} \right] \tilde{\rho}_{31} - A' \tilde{\rho}_{21} &= -G_2 \tilde{\rho}_{32} - G_3 \tilde{\rho}_{33}, \end{aligned} \quad (6.2)$$

where the indices 1, 2, and 3 now number the "Stark" states of the atom in a constant field with the wave functions of the three-level system

$$\begin{aligned} \tilde{\phi}_1 &= \phi_1, \\ \tilde{\phi}_2 &= \phi_2 \cos \beta/2 + \phi_3 \sin \beta/2, \\ \tilde{\phi}_3 &= -\phi_2 \sin \beta/2 + \phi_3 \cos \beta/2, \end{aligned} \quad (6.3)$$

and, in addition, we introduce the notation

$$\tilde{v} = v \cos^2 \beta, \quad v \equiv 2\Phi,$$

$$\begin{aligned}
A' &= \cos \beta/2 \sin \beta/2 \frac{A_2 - A_3}{2} = \frac{1}{2} \operatorname{tg} \beta \frac{\bar{A}_2 - \bar{A}_3}{2}, \\
\bar{A}_2 &= A_2 \cos^2 \beta/2 + A_3 \sin^2 \beta/2, \quad A_2 = \Gamma_{22} + A_{21}, \\
\bar{A}_3 &= A_2 \sin^2 \beta/2 + A_3 \cos^2 \beta/2, \quad A_3 = \Gamma_{33} + A_{31}, \\
\bar{Q}_2 &= Q_2 \cos^2 \beta/2 + Q_3 \sin^2 \beta/2, \\
\bar{Q}_3 &= Q_2 \sin^2 \beta/2 + Q_3 \cos^2 \beta/2, \\
G_2 &= G \cos \beta/2, \quad G_3 = -G \sin \beta/2, \\
\bar{\omega}_{21} &= \omega_0 - \bar{\omega}_{32}/2, \quad \bar{\omega}_{31} = \omega_0 + \bar{\omega}_{32}/2, \quad \omega_0 = (\omega_{21} + \omega_{31})/2, \\
\bar{\omega}_{32} &= (\omega_{32}^2 + 4V^2)^{1/2} = \omega_{32}/\cos \beta,
\end{aligned} \tag{6.4}$$

and for  $\cos^2 \beta/2$  and  $\sin^2 \beta/2$  there exist the convenient relations

$$\cos^2 \beta/2 = \frac{\bar{\omega}_{32} + \omega_{32}}{2\bar{\omega}_{32}}, \quad \sin^2 \beta/2 = \frac{\bar{\omega}_{32} - \omega_{32}}{2\bar{\omega}_{32}}. \tag{6.5}$$

It is natural that using Eqs. (6.1)–(6.5) leads to the same result for the line shape as in the spherical basis [see Eqs. (3.6) and (3.7)]:

$$I(\omega) = \frac{1}{\pi} \left\langle \operatorname{Im} \frac{\Omega' + R_1 + i(\gamma_3 + R_2)}{V^2 - (\Omega + i\gamma_2)(\Omega' + i\gamma_3)} \right\rangle, \tag{6.6}$$

$$\gamma_2 = \frac{\Gamma_{11} + A_2 + \nu}{2}, \quad \gamma_3 = \frac{\Gamma_{11} + A_3 + \nu}{2}, \tag{6.7}$$

$$R_1 = V \operatorname{Re}(\bar{\rho}_{32}^0/\rho_{22}^0), \quad R_2 = V \operatorname{Im}(\bar{\rho}_{32}^0/\rho_{22}^0), \tag{6.8}$$

where  $\Omega \equiv \omega - \omega_{21}$  and  $\Omega' \equiv \omega - \omega_{31}$  are the shifts from the frequency of the allowed ( $\omega_{21}$ ) and the “forbidden” ( $\omega_{31}$ ) transitions. Here

$$R_1 = \frac{2\omega_{32}}{A_2 + A_3} R_2, \quad R_2 = V^2 \frac{A_2 Q_3 - A_3 Q_2}{A_3 Q_2 \Gamma + (Q_2 + Q_3)(2V^2 + \nu\Gamma)}, \tag{6.9}$$

$$\Gamma = 2 \frac{\omega_{32}^2}{A_2 + A_3} + 2\nu + \frac{A_2 + A_3}{2}.$$

In the frequently arising case

$$\gamma_2, \gamma_3 \ll \omega_{32}, \tag{6.10}$$

it is possible to represent the total line shape in the form of a sum of two Lorentzian line shapes corresponding to the allowed and forbidden ( $A_{21} \gg A_{31}$ ) components of the line:

$$\begin{aligned}
I(\omega) &= \frac{1}{\pi} \left\langle \left[ \frac{(\Gamma_2/2)(\cos^2 \beta/2 - \delta) - \Omega_a \delta'}{\Omega_a^2 + \Gamma_2^2/4} \right. \right. \\
&\quad \left. \left. + \frac{(\Gamma_3/2)(\sin^2 \beta/2 + \delta) + \Omega_f \delta'}{\Omega_f^2 + \Gamma_3^2/4} \right] \right\rangle, \tag{6.11}
\end{aligned}$$

where

$$\begin{aligned}
\Omega_a &= \omega - \bar{\omega}_{21}, \quad \Omega_f = \omega - \bar{\omega}_{31}, \\
\Gamma_2 &= \Gamma_{11} + \bar{A}_2 + \nu = 2(\gamma_2 \cos^2 \beta/2 + \gamma_3 \sin^2 \beta/2), \\
\Gamma_3 &= \Gamma_{11} + \bar{A}_3 + \nu = 2(\gamma_2 \sin^2 \beta/2 + \gamma_3 \cos^2 \beta/2).
\end{aligned} \tag{6.12}$$

The NIEF contribution to the line shape is determined by the parameters

$$\begin{aligned}
\delta &= R_1/\bar{\omega}_{32}, \\
\delta' &= \frac{2V^2 A_2}{\bar{\omega}_{32}^3} \left[ \frac{1}{2} \left( 1 - \frac{A_3}{A_2} \right) - \frac{R_1}{\omega_{32}} \left( 1 + \frac{A_3}{A_2} \frac{2V^2 + \omega_{32}^2}{2V^2} \right) \right].
\end{aligned} \tag{6.13}$$

$$\tag{6.14}$$

For  $\Omega_a, \Omega_f \gg \Gamma_2, \Gamma_3$  the terms proportional to  $\delta'$  cancel and the NIEF contribution depends only on  $\delta$ . In addition to this, the terms with  $\delta'$  disappear at the peaks of the components. The amplitudes of the allowed and forbidden peaks are

$$I_a^{(0)} = \frac{\cos^2 \beta/2}{\pi \Gamma_2/2}, \quad I_f^{(0)} = \frac{\sin^2 \beta/2}{\pi \Gamma_3/2}. \tag{6.15}$$

In the limit  $V \rightarrow \infty$  or  $V \rightarrow 0$  NIEF is absent, i.e.,  $\delta, \delta' \rightarrow 0$ . Therefore if the terms with  $\delta$  and  $\delta'$  in Eq. (6.11) are discarded, we obtain the intensities of the components in the purely spherical or parabolic bases with probabilities  $\bar{A}_2$  and  $\bar{A}_3$  (6.4). Taking NIEF into account leads to a redistribution of the intensities:  $I_a^{(0)}$  decreases while  $I_f^{(0)}$  grows. The relative contribution of NIEF to the forbidden component can be characterized by the quantity

$$\xi = \frac{\delta}{\sin^2 \beta/2}. \tag{6.16}$$

Let us consider for simplicity the case  $A_3 = 0, A_2 = 0$ . Then

$$\xi = \frac{Q_3}{Q_2 + Q_3} \frac{4V^2}{(2V^2 + \nu\Gamma) [(1 + 4V^2/\omega_{32}^2)^{1/2} - 1]}. \tag{6.17}$$

In weak fields, for  $V \ll \omega_{32}, (\nu\Gamma)^{1/2}$ ,

$$\xi = \frac{Q_3}{Q_2 + Q_3} \frac{A}{\nu} \frac{1}{1 + A(4\nu + A)/4\omega_{32}^2},$$

i.e., the magnitude of the contribution of NIEF depends on the ratio of the radiative and collisional widths. For ions with small nuclear charge  $Z$  under real conditions we have  $\xi \ll 1$ , but with increasing  $Z$  this contribution grows since  $A \sim Z^4$ , but  $\nu \sim Z^{-2}$ . For  $V \gg \omega_{32}, (\nu\Gamma)^{1/2}$  the relative contribution of NIEF falls off with the field as  $V^{-1}$ :

$$\xi = \frac{Q_3}{Q_2 + Q_3} \frac{\omega_{32}}{V} \ll 1,$$

but the amplitudes of the forbidden and allowed components (6.15) are comparable ( $\delta \rightarrow 0, \Gamma_2 \rightarrow \Gamma_3, \cos^2 \beta/2 \rightarrow 1/2$ ).

Thus, the NIEF contribution turns out to be substantial for  $Z \sim 20\text{--}30$ , while condition (6.10), more exactly  $\Delta\Gamma = \Gamma_2 - \Gamma_3 \ll \bar{\omega}_{32}$ , is satisfied all the way up to  $Z \leq 50$ .

From a comparison of system of equations (3.6), (3.7) with system of equations (6.2) it is clear that when we go to the parabolic basis the field terms fall out and the frequencies  $\omega_{ij}$  are simultaneously replaced by the field-perturbed frequencies  $\bar{\omega}_{ij}$ , complicating the form of the collision operator. In the case met in practice  $\gamma \ll \omega_{32}$  and the parameter  $\delta$  can be represented in the form (for  $A_3 = 0, A = A_2$ )

$$\delta = \frac{Q_3}{Q_2 + Q_3} \frac{\omega_{32}}{\bar{\omega}_{32}} \frac{V^2}{V^2 + \nu\omega_{32}^2/A}. \tag{6.18}$$

This result also follows from the simplified system

$$\begin{aligned}
\bar{A}_2 \bar{\rho}_{22} - \bar{\nu}(\bar{\rho}_{33} - \bar{\rho}_{22}) &= \bar{Q}_2, \\
\bar{A}_3 \bar{\rho}_{22} + \bar{\nu}(\bar{\rho}_{33} - \bar{\rho}_{22}) &= \bar{Q}_3,
\end{aligned} \tag{6.19}$$

$$[i(\bar{\omega}_{21} - \omega) + (\bar{A}_2 + \nu)/2] \bar{\rho}_{21} = -\bar{G}_2 \bar{\rho}_{22},$$

$$[i(\bar{\omega}_{31} - \omega) + (\bar{A}_3 + \nu)/2] \bar{\rho}_{31} = -\bar{G}_3 \bar{\rho}_{33},$$

which arises upon discarding the terms in  $A'$  and the nondia-

gonal elements  $\tilde{\rho}_{23}$  and  $\tilde{\rho}_{32}$  in Eqs. (6.2). This has to do with the fact that these terms are small in the parabolic basis in comparison with the population difference  $\Delta\tilde{\rho} = \tilde{\rho}_{33} - \tilde{\rho}_{22}$  with small proportionality parameter  $\gamma/\omega_{32}$ :

$$\operatorname{Re} \tilde{\rho}_{32}/\Delta\tilde{\rho} \sim \frac{V}{\omega_{32}} \frac{A(A+4\nu)}{\tilde{\omega}_{32}^2}, \quad (6.20)$$

$$\operatorname{Im} \tilde{\rho}_{32}^0/\Delta\tilde{\rho} \sim \left( \frac{V}{\omega_{32}} \right) (A/\tilde{\omega}_{32}),$$

while in the spherical basis only the imaginary part of  $\tilde{\rho}_{32}^0$  remains small:

$$\operatorname{Re} \tilde{\rho}_{32}^0/\Delta\rho \sim V/\omega_{32}, \quad (6.21)$$

$$\operatorname{Im} \tilde{\rho}_{32}^0/\Delta\rho \sim (V/\omega_{32}) (A/\omega_{32}).$$

If we neglect the first pair of Eqs. (6.19), then we obtain the conventional result for the theory of spectral line broadening, which is based solely on a consideration of the evolution operator<sup>1</sup> and is in essence equivalent to just the second pair of Eqs. (6.19) (for the polarizations  $\tilde{\rho}_{21}$  and  $\tilde{\rho}_{31}$ ). In this case the populations  $\tilde{\rho}_{22}$  and  $\tilde{\rho}_{33}$  are assumed to be identical, and the nonlinear interference effects disappear completely. Precisely such an approach, based on the assumption of equal occupancy of levels 2 and 3, is used in Ref. 5 to calculate the intensities of the forbidden components in the spectra of atoms of helium, lithium, and copper. It thereby becomes clear that NIEF is due to the inequality of the populations of the emitting levels in any basis.

## 7. NUMERICAL RESULTS FOR THE LINE SHAPE AND THE RESCATTERING FUNCTION

### a. The line shape

The calculations were carried out for the transitions  $2^1S-4^1P$  and  $2^1S-4^1P$  of helium-like ions of aluminum and nickel:  $\text{Al}^{11+}$  and  $\text{Ni}^{26+}$ . The necessary atomic parameters, given below, were obtained in the Hartree-Fock approximation, taking into account the interaction of configurations (Cowan's program<sup>23</sup>):

$$\begin{array}{cc} \text{Al}^{11+} & \text{Ni}^{26+} \\ A_{21}=2,3 \cdot 10^{11} \text{ s}^{-1} & A_{21}=5,1 \cdot 10^{12} \text{ s}^{-1} \\ \Gamma_{22}+A_{21}=4,1 \cdot 10^{12} \text{ s}^{-1} & \Gamma_{22}+A_{21}=8,3 \cdot 10^{13} \text{ s}^{-1} \\ \omega_{32}=4 \cdot 10^{14} \text{ s}^{-1} & \omega_{32}=1,3 \cdot 10^{15} \text{ s}^{-1} \end{array}$$

Here the indices 1, 2, and 3 denote respectively the levels  $2^1S$ ,  $4^1P$ , and  $4^1D$ . Note that the energy difference changes sign at the value of the nuclear charge  $Z \approx 22$ .

This behavior the dependence of  $\omega_{32}(Z)$  was confirmed by the calculations of Vainstein and Safronova<sup>24</sup> by means of an expansion in the parameter  $1/Z$ . The absolute values of the energies of the excited states of the helium-like ions, obtained by different authors,<sup>23-26</sup> agree to within four or five significant figures. However, the differences in energy of near-lying levels are determined with considerably less accuracy. For example, the values of  $\omega_{32}$  according to the data of Refs. 23 and 24 differ for Al by  $\sim 30\%$ , and for Ni this discrepancy stands at  $\sim 10\%$ . In our calculations we gave preference to Cowan's program,<sup>23</sup> which provides the possibility of obtaining the entire set of atomic data within a single approach. Nevertheless, it should be borne in mind that our final results (the line shape, the rescattering function) have for the most part a qualitative character and have as their goal to demonstrate first of all the role of NIEF.

Evaluation of the collisional width gives<sup>7</sup>

$$\nu[\text{s}^{-1}] = 3,1 \cdot 10^{-5} N_e [\text{cm}^{-3}] (Z-1)^{-2} (T_e[\text{eV}])^{-1/2}.$$

The matrix element  $V$  of the interaction of the atom with the field for the mean Holtsmark field intensity is usually taken to be equal to

$$V[\text{s}^{-1}] = 30 (N_e [\text{cm}^{-3}])^{1/2}.$$

This quantity corresponds to the value of the dipole moment averaged over the magnetic quantum numbers of the states  $4^1P_1$  and  $4^1D_2$ . This approximation gives a simplified picture of the Stark splitting (e.g., The  $4^1F_3$  Stark states, which substantially influence the magnitude of  $\omega_{32}$ , are not taken into account) and can be justified only as the first stage of calculations which have a model character.

The radiative transition  $3 \rightarrow 1$  ( $4^1D \rightarrow 2^1S$ ) is dipole-forbidden, but the probability of the corresponding quadrupole transition is negligibly small in comparison with  $A_{21}:A_{31} = 2,6 \cdot 10^7 \text{ s}^{-1}$  for  $\text{Al}^{11+}$  and  $A_{31} = 1,9 \cdot 10^9 \text{ s}^{-1}$  for  $\text{Ni}^{26+}$ , so that the quadrupole transition plays no substantial role in determining the line shape.

Figure 3a depicts the line shape of the spectral doublet  $2^1S-4^1P$ ,  $4^1D$  of the helium-like ion of Al in a plasma with an electron density of  $N_e = 10^{20} \text{ cm}^{-3}$  and temperature  $T_i = T_e = 350 \text{ eV}$ , calculated according to Eq. (6.11).<sup>6)</sup> The pumping at the levels 2 and 3 was assumed to be identical:  $Q_1 = Q_3$ . The given figure corresponds to line shape not averaged over the velocities and the microfield distribution: 1—the total line shape, 2—the NIEF contribution, and 3—the line shape without account of NIEF. It is clear that

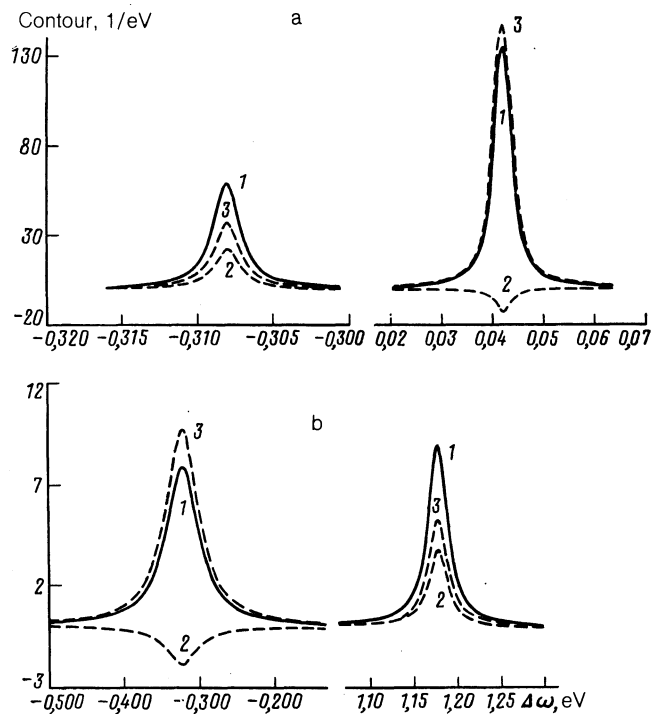


FIG. 3. a) Line shape of the spectral doublet  $2^1S-4^1P$ ,  $4^1D$  of the helium-like aluminum ion, not Doppler-averaged. The magnitude of the field intensity corresponds to the average Holtsmark field.  $N_e = 10^{20} \text{ cm}^{-3}$ ,  $T_i = T_e = 350 \text{ eV}$ ,  $Q_2 = Q_3$ . Curve 1) the total line shape, curve 2) the NIEF contribution, curve 3) the line shape without NIEF; b) line shape of the same transition as in Fig. 3a, but for the helium-like nickel ion at  $N_e = 10^{21} \text{ cm}^{-3}$  and  $T_e = T_i = 500 \text{ eV}$ . Notation the same as in Fig. 3a.

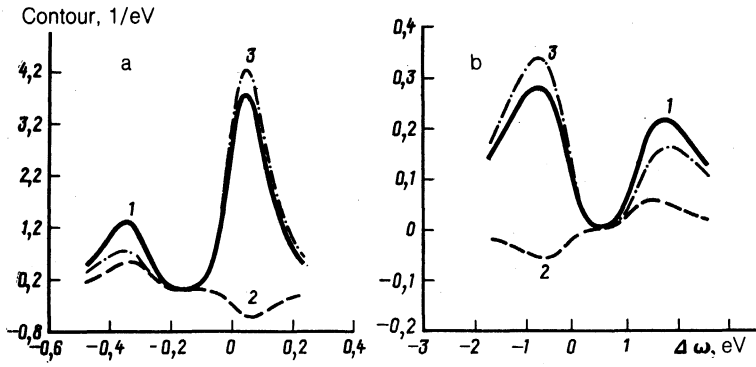


FIG. 4. a) Same line shape as in Fig. 3a, but averaged over the microfields and the velocities of the emitters; b) same line shape as in Fig. 3b, but averaged over the microfields and the velocities of the emitters.

NIEF leads to a decrease in the intensity of the allowed component (the right peak) and some increase in the forbidden component (the left peak). Averaging over the microfields and velocities makes the NIEF contribution still more significant, as Fig. 4a shows.

Figure 3b shows the analogous line shape for the helium-like ion of Ni for an electron density  $N_e = 10^{21} \text{ cm}^{-3}$  and temperature  $T_i = T_e = 500 \text{ eV}$ . Under the given conditions before averaging over the velocities and the fields with NIEF taken into account the forbidden component has a higher peak than the allowed one, but after averaging (Fig. 4b) the allowed component becomes more intense (although insignificantly so, by no more than a factor of 1.5).

It is clear that in all of the calculated cases the NIEF contribution substantially varies the ratio between the intensities of the allowed and forbidden components of the line.

### b. The rescattering function

Using this theory we carried out calculations of the incoherent part of the frequency redistribution function

$R(\omega, \omega')$  for the helium-like ions of Al and Ni (the transitions and constants the same as in the calculations of the emission contour) in a plasma at different densities. Figures 5a and b depict the function  $R(\omega, \omega')$  at  $T_e = 350 \text{ eV}$  and  $N_e = 10^{20} \text{ cm}^{-3}$  for the helium-like aluminum ion in the mean Holtsmark microfield intensity (Fig. 5a, with NIEF taken into account; Fig. 5b, with NIEF not taken into account). Figures 5c and d depict the rescattering function for the helium-like nickel ion at  $T_e = 500 \text{ eV}$  and  $N_e = 6.5 \cdot 10^{22} \text{ cm}^{-3}$  (c) with NIEF, d) without NIEF). It is clear that NIEF is manifested in the redistribution of intensity between the peaks. At large fields ( $N_e \geq 10^{22} \text{ cm}^{-3}$ ) NIEF leads to a strong suppression of the peaks corresponding to the condition  $\Omega\Omega' = -V^2$  according to the approximate formula (5.42). At small fields ( $N_e \leq 10^{21} \text{ cm}^{-3}$ ) the role of NIEF is at its most significant for the value of the redistribution function  $Q_{\text{inc}}(\Omega, \Omega')$  at the point  $\Omega = \Omega' = \omega_{32}$  according to Eq. (5.43) (cf. Figs. 5a and b). In this limit the parameter that characterizes the contribution of NIEF to the redistribution function, analogous to the

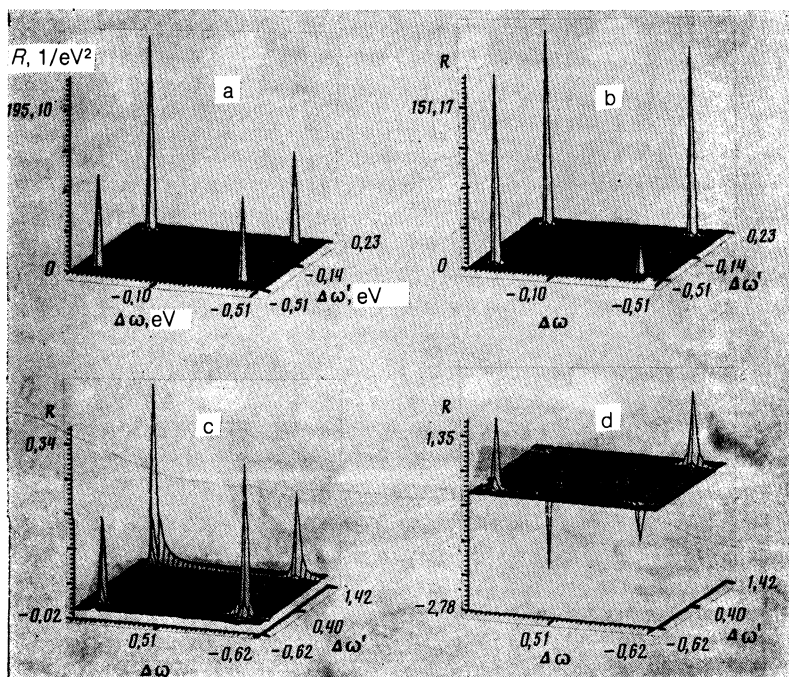


FIG. 5. Frequency redistribution function  $R(\omega, \omega')$  for the spectral doublet  $2^1S-4^1P, 4^1D$  of the helium-like aluminum ion, but averaged over the fields and velocities at  $T_e = 350 \text{ eV}$ ,  $N_e = 10^{20} \text{ cm}^{-3}$  (a, b) and the helium-like nickel ion at  $T_e = 500 \text{ eV}$ ,  $N_e = 6.5 \cdot 10^{22} \text{ cm}^{-3}$  (c, d); a) and c) the form of the function  $R(\omega, \omega')$  with NIEF taken into account; b) and d) the form of the function  $R(\omega, \omega')$  with NIEF not taken into account.

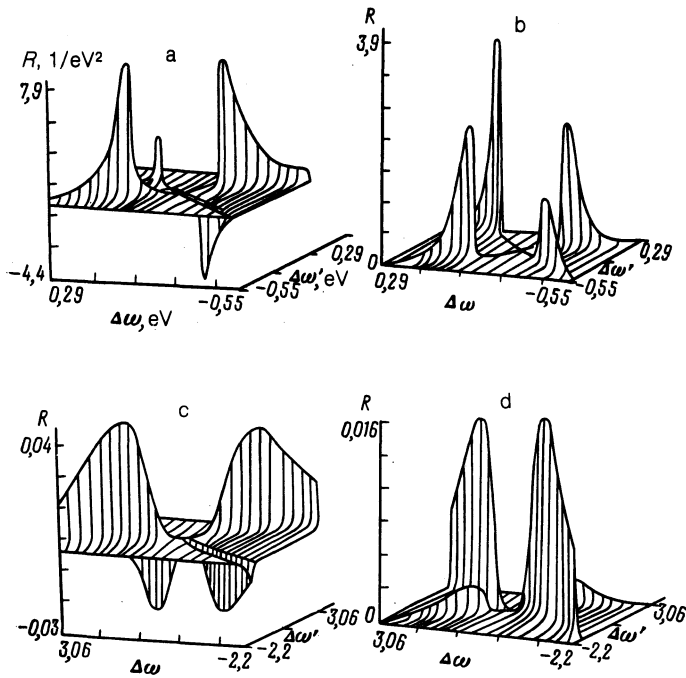


FIG. 6. The frequency redistribution function  $R(\omega, \omega')$  averaged over the microfields for the spectral doublet  $2^1S-4^1P, 4^1D$  of the helium-like aluminum ion at  $T_e = 350$  eV,  $N_e = 10^{20}$  cm $^{-3}$  (a, b) and the helium-like nickel ion at  $T_e = 500$  eV,  $N_e = 6.5 \cdot 10^{22}$  cm $^{-3}$  (c, d); a) and c) the form of the function  $R(\omega, \omega')$  with NIEF taken into account; b) and d) the form of the function  $R(\omega, \omega')$  with NIEF not taken into account.

parameter  $\xi$  [see Eqs. (6.16) and (6.17)] can be written in the form

$$\xi \sim \left[ \frac{(v/A) \omega_{32}^2}{\omega_{32}^2 + AV^2/v} \right]^{-1}.$$

Here we have  $v \ll A$ , and the larger  $\xi$  is, the larger is the NIEF contribution.

Note that at relatively small fields ( $V \ll 10^{19}$  cm $^{-3}$ ) negative values of the rescattering function, calculated without account of NIEF, can exist in the region  $\omega \approx \omega' \approx \omega_{32}$  [see formulas (5.44) and (5.45) and Fig. 5d]. This once again emphasizes the necessity of taking NIEF into account.

In the region of higher fields ( $V \gg \omega_{32}$ ,  $V \gg v \gg A$ ) the contribution of NIEF to the peaks corresponding to  $\Omega \Omega' = -V^2$  is also determined by the quantity  $v/A$ , which in this limit is much smaller than unity.

Note that the values of the incoherent part of the redistribution function shown in Figs. 5a and c were calculated for the mean Holtmark electric microfield intensity. The true redistribution function is obtained according to Eqs. (5.23) and (5.33) by averaging over the microfield distribution and over the velocities of the absorbing ions. Figure 6 shows the rescattering function for helium-like aluminum (nickel) ions, averaged over the microfields, for conditions analogous to those of Fig. 5 (Fig. 6a and c—the total function, Fig. 6b and d—the function without NIEF taken into account). Qualitatively, as in the calculation of the emission contour, the role of NIEF is preserved after averaging over the microfields and velocities. During averaging there is the possibility of redistribution between the positive and negative regions of the values of the rescattering function, calculated without account of NIEF (cf. Fig. 5a and Fig. 6a). In radiative transfer theory frequency use is made of the rescattering function  $R(\omega, \omega')$  averaged over the angles of the incident and scattered photons (cf. Ref. 13) and even the thermal motion of the ions. In the present paper we do not present the results of this averaging.

Note that the high sensitivity of the frequency redistribution function of the photons to NIEF in an MCI plasma can be used as a diagnostic for an optically thin plasma irradiated by resonant radiation which is then scattered by the absorbing ions. As was already noted, the function  $R(\omega, \omega')$  is more sensitive to the role of NIEF than the emission contour, which characterizes the emissivity of the optically thin MCI plasma.

In the case of an optically thick plasma transport of resonant radiation with the Stark microfields taken into account possesses a number of qualitative features associated with the nature of the redistribution function and the influence of NIEF on the magnitude of the function  $R(\omega, \omega')$ . In the case of small values of  $v/A$  the redistribution function is almost coherent [see Eq. (5.35)]; however, the multiplier of the  $\delta$ -function does not coincide with the line shape  $\phi(\omega)$ , and at the point  $\Omega = \omega_{32}$  this difference can be large, proportional to the parameter  $V^2/vA \gg 1$ . At large values of  $v/A$  the incoherent component plays the main role in the radiative transfer. However, because of the role of NIEF noted above the regime of complete frequency redistribution (CFR, see Ref. 13) is not realized in this limit, which means that  $R(\omega, \omega') \approx \phi(\omega)\phi(\omega')$  and the spectral density of the excited atoms  $N(\omega) \approx N\phi(\omega)$ , where  $N$  is the total density of excited particles. This difference from CFR is connected with the fact that the average of the product of the Stark microfields does not coincide with the product of the averages:  $\langle \phi(\omega)\phi(\omega') \rangle \neq \langle \phi(\omega) \rangle \langle \phi(\omega') \rangle$ , and the NIEF contribution strongly influences the redistribution of intensities of the peaks of the rescattering function.

In conclusion we express our thanks to A. A. Panteleev and V. A. Roslyakov for their interest in this work and helpful discussions.

#### APPENDIX

To describe the atomic subsystem we introduce four Green's functions:

$$\begin{aligned}
iG^{--}(x, \xi; x', \xi') &= \langle T(\hat{\psi}(x, \xi)\hat{\psi}^+(x', \xi')) \rangle, \\
iG^{++}(x, \xi; x', \xi') &= \langle \tilde{T}(\hat{\psi}(x, \xi)\hat{\psi}^+(x', \xi')) \rangle, \\
iG^{+-}(x, \xi; x', \xi') &= \langle \hat{\psi}(x, \xi)\hat{\psi}^+(x', \xi') \rangle, \\
iG^{-+}(x, \xi; x', \xi') &= -\langle \hat{\psi}^+(x, \xi)\hat{\psi}(x', \xi') \rangle.
\end{aligned} \tag{A1}$$

Here  $\psi(x, \xi)$  is the particle field operator in the Heisenberg representation, and  $T$  and  $\tilde{T}$  are the chronological and antichronological ordering operators. In the interaction representation the following expansion is valid for the operator  $\hat{\psi}(x, \xi)$

$$\hat{\psi}(x, \xi) = \frac{1}{V^{1/2}} \sum_{\mathbf{p}, j} \hat{a}_{\mathbf{p}, j} \tilde{\phi}_{\mathbf{p}, j}(x, \xi, t). \tag{A2}$$

Here  $V$  is the volume of the system,  $\hat{a}_{\mathbf{p}, j}$  is the annihilation operator of the atomic particles possessing momentum  $\mathbf{p}$  in the state  $j$ . For definiteness we assume Fermi statistics. The wave functions of the atomic particles in the external electric field  $\mathbf{F}$  satisfy the equation

$$i \frac{\partial}{\partial t} \tilde{\phi}_j = \left( -\frac{\Delta_{\mathbf{r}}}{2M} - \hat{\mathbf{d}}_{\xi} \mathbf{F} + H_0(\xi) \right) \tilde{\phi}_j(x, \xi, t), \tag{A3}$$

where  $H_0(\xi)$  is the Hamiltonian of the atomic subsystem which determines the eigenstates of the system  $\phi_j(\xi)$  with energies  $\hbar\omega_j$ :

$$H_0(\xi) \phi_j(\xi) = \hbar\omega_j \phi_j(\xi). \tag{A4}$$

the function  $\phi_j$  is found from Eq. (A3) with the initial conditions ( $t \rightarrow -\infty$ ):

$$\tilde{\phi}_j \xrightarrow{t \rightarrow -\infty} \tilde{\phi}_j(\xi) \exp(i\mathbf{p}\mathbf{r} - i(\omega_j + \varepsilon_{\mathbf{p}})t). \tag{A5}$$

The projections of the Green's functions on the wave functions of Eq. (A4), i.e., onto the original spherical basis, are obviously equal to

$$G_{ij}^{aa'}(x, x') = \int \phi_j^*(\xi) G^{aa'}(x, \xi; x', \xi') \phi_i(\xi') d\xi d\xi'. \tag{A6}$$

We restrict ourselves to the dipole approximation in the description of the interaction of the atomic system with the Stark field  $\mathbf{F}$  with quantized electromagnetic radiation field. In this case we use the operator of the intensity of the transverse quantized field in the interaction representation  $\hat{E}_i(x)$  in the form

$$\begin{aligned}
\hat{E}_i(x) &= \sum_{\mathbf{k}, \lambda} (2\pi\omega_{\mathbf{k}}/V)^{1/2} [\mathbf{e}_i(\mathbf{k}, \lambda) \hat{b}_{\mathbf{k}, \lambda} \exp(-i\omega_{\mathbf{k}}t + i\mathbf{k}\mathbf{r}) \\
&\quad - \mathbf{e}_i^*(\mathbf{k}, \lambda) \hat{b}_{\mathbf{k}, \lambda}^{\dagger} \exp(i\omega_{\mathbf{k}}t - i\mathbf{k}\mathbf{r})].
\end{aligned} \tag{A7}$$

Here  $\hat{b}_{\mathbf{k}, \lambda}$  is the annihilation operator of the photon with wave vector  $\mathbf{k}$  and polarization  $\lambda$ ,  $\mathbf{e}_i(\mathbf{k}, \lambda)$  is the unit vector in the direction of the field,  $\omega_{\mathbf{k}}$  is the frequency of the photon with wave vector  $\mathbf{k}$ , and  $\omega_{\mathbf{k}} = c|\mathbf{k}|$ .

In analogy with Eq. (A1) it is customary to introduce the Green's function for the photon subsystem. For example,

$$iD_{jk}^{-+}(x, x') = \langle \hat{E}_j(x') \hat{E}_k(x) \rangle, \tag{A8}$$

where the operator  $\hat{E}_j$  is already in the Heisenberg representation.

Broadening of the atomic subsystem by electrons is characterized in the Fourier representation by the matrix element of the interaction potential  $V_{21}(\mathbf{q}, \omega)$ .

The retarding and accelerating Green's functions, e.g., for the atomic system, are defined by the relations

$$\begin{aligned}
G_{ik}^R &= G_{ik}^{--} - G_{ik}^{-+}, \\
G_{ik}^A &= G_{ik}^{-+} - G_{ik}^{++}.
\end{aligned} \tag{A9}$$

Transforming from the coordinates  $x, x'$  to the coordinates

$$\mathbf{R} = \frac{\mathbf{r} + \mathbf{r}'}{2}, \quad \boldsymbol{\rho} = \mathbf{r} - \mathbf{r}', \quad T = \frac{t + t'}{2}, \quad \tau = t - t'$$

and making use of the Fourier transformation of the Green's functions with respect to  $\boldsymbol{\rho}$  and  $\tau$ , we arrive at the functions  $G_{ik}^{\alpha\alpha'}(\omega, \mathbf{p}, \mathbf{R}, T)$ , which figure in the main text.

- <sup>1</sup> The transition 3 → 1 may be forbidden.
- <sup>2</sup> The variable  $\bar{\tau}_{21}$  determines the time-dependent part of the rescattered power, which disappears upon averaging over a period. For its physical meaning see Ref. 10 and 8.
- <sup>3</sup> The main definitions are given in the Appendix.
- <sup>4</sup> Note that the atomic density matrix used in the present section has no simple relation to the density matrix of the compound system which figures in Sec. 2.
- <sup>5</sup> The coherent term can be distinguished only when there is no decay of the lower level:  $\Gamma_{11} \rightarrow 0$ .
- <sup>6</sup> The quantities  $\Delta\omega = \omega - \omega_{21}$  and  $\Delta\omega' = \omega' - \omega_{21}$  are plotted along the horizontal axes in Figs. 3–6.
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