Narrowing of nonlinear resonances in a collisional plasma

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The profile of a Bennett structure induced by an electromagnetic field in the velocity distribution of the particles and the spectrum of the probe field with allowance for the velocity dependence of the diffusion tensor have been calculated for the first time. In the case of Coulomb scattering of the plasma particles, the decrease in the collision rate with increasing particle velocity may lead to a narrowing of the Bennett structure or the probe field spectrum as the deviation of the wave frequency from the resonant value increases. The renormalization method is used to calculate the resonance's shape and width. We also provide estimates demonstrating that with an argon laser the effect reaches 10% and can be observed. The calculations done in this paper can be used to measure the velocity dependence of the diffusion tensor. [S1063-7761(96)00802-3]

1. INTRODUCTION

One way of studying collision processes in a gas in a contact-free manner is to investigate the resonant interaction of the gas and a traveling electromagnetic wave.[1,2] If the wave frequency ω is close to the Bohr frequency $\omega_{mn}=(E_m-E_n)/\hbar$ of the transition between states $|m\rangle$ and $|n\rangle$, then due to the Doppler effect the particles that interact most effectively with the wave are those for which $\mathbf{k}\cdot\mathbf{v}=\omega-\omega_{mn}$, where $\mathbf{k}$ is the wave vector of the electromagnetic wave, and $\mathbf{v}$ is the particle velocity. Such particles are involved in transitions from state $|m\rangle$ to state $|n\rangle$ and back. As a result the population of state $|m\rangle$ (or $|n\rangle$), considered as a function of the particle velocity, acquires a narrow peak or dip (the Bennett dip[3]). The shape of this feature largely depends on the collision rate, and a change in it occurs, for one thing, because during scattering the particles change their velocity and consequently may leave the resonance sooner than quenching of the states $|m\rangle$ and $|n\rangle$ can take place.

Experimentally the distribution of the particles over the quantum states can be studied by directing another electromagnetic field of frequency ω_μ and low intensity (the probe field), which leaves the distribution of the particles over the states practically unchanged. The power absorbed from the probe wave is proportional to the number of particles in resonance with the wave, i.e., moving with velocities that meet the condition $\mathbf{k}_\mu\cdot\mathbf{v}=\omega_\mu-\omega_{mn}$. Hence by changing the frequency ω_μ of the probe wave we scan, so to speak, the velocity distribution of particles that are in state $|m\rangle$ (or $|n\rangle$).[4]

This paper examines the case when the collision process can be described as a diffusion in velocity space. The collision integral in the kinetic equation is replaced by a second-order differential operator acting on the velocity distribution function. Such a description of scattering is possible, for instance, for ions in a plasma with Coulomb scattering[5,6] or for heavy particles in a light-particle buffer gas.[7] In an equilibrium plasma or in an equilibrium velocity distribution of the buffer gas particles, the differential operator describing diffusion is proportional to

$$\frac{\partial}{\partial v_\alpha}\ \Phi_{\alpha\beta}(\mathbf{v})\left(\frac{\partial}{\partial v_\beta}+\frac{2v_\beta}{v_T^2}\right),$$

where v_T is the thermal velocity of the particles investigated, and $\Phi_{\alpha\beta}(\mathbf{v})$ is the diffusion tensor. Usually the kinetic equation is solved in an approximation in which the diffusion tensor is velocity-independent (see Ref. 5). In Ref. 6 the kinetic equation is solved with allowance for the velocity dependence of the diffusion tensor but in the linear approximation in the strength of the wave field. The velocity distribution of the particles has been briefly discussed in Ref. 8. In the present paper we do a detailed calculation of the first nonlinear approximation for the shape of the Bennett dip (the particle distribution averaged over the particle velocities that are transverse in relation to the direction of wave propagation) and the work of the probe field in the case when the diffusion tensor is velocity-dependent.

When solving the kinetic equation in the first-order perturbation approximation, we can easily write the expressions for the Bennett dip and the probe field spectrum in the form of quadruple and quintuple integrals, respectively, but such expressions are difficult to analyze and use in quantitative calculations. In this paper we suggest approximations in which these expressions are reduced to simple integrals containing only one component of the diffusion tensor. We also estimate the magnitude of the effect in conditions typical of an argon laser in which the low-temperature plasma is the active medium, and give the results of numerical calculations that support the approximations.

In Sec. 2 we introduce the equations for the density matrix and discuss the velocity dependence of the components of the diffusion tensor of a test ion in an equilibrium plasma. Section 3 is devoted to calculating the first-order correction to the distribution function of particles that are in a definite quantum state, the correction being related to allowing for the interaction of a two-level system in state $|m\rangle$ or $|n\rangle$ and an electromagnetic wave. In Sec. 4 we calculate the probe field spectrum, an observable quantity, i.e., the power absorbed from the probe wave as a function of the mismatch between the wave's frequency and the resonant frequency.

2. THE FOKKER–PLANCK EQUATION

Let us examine a gas consisting of identical particles. The Maxwellian velocity distribution is normalized to unity:

$$W(\mathbf{v})=\frac{1}{(\sqrt{\pi}v_T)^3}\exp(-\mathbf{v}^2/v_T^2),$$

$$v_T=\sqrt{\frac{2k_BT}{M}},\qquad \int d\mathbf{v}W(\mathbf{v})=1, \tag{1}$$

where T is the temperature of the gas, M is the mass of the particles, and k_B is Boltzmann's constant. We take two excited particle states $|m\rangle$ and $|n\rangle$ with energies E_m and E_n (to be specific we assume $E_m>E_n$) and the resonant interaction of the given two-level system with a traveling electromagnetic wave in the form

$$\mathbf{E}(t,\mathbf{r})=\tfrac{1}{2}(\mathbf{E}_0\exp\{-i(\omega t-\mathbf{kr})\}+\mathbf{E}_0^*\exp\{i(\omega t-\mathbf{kr})\}), \tag{2}$$

i.e., $\Omega=\omega-\omega_{mn}\ll\omega$, where $\omega_{mn}=(E_m-E_n)/\hbar$.

Assuming that the interaction of the particle and wave is of the dipole type, in the resonance approximation we have the following quantum Fokker–Planck equations for the density matrix:[5]

$$\begin{cases}\left(\frac{\partial}{\partial t}+(\mathbf{v}\nabla_\mathbf{r})+\Gamma_j\right)\rho_j=Q_jW(\mathbf{v})+\nu\hat{\mathscr{V}}\rho_j\\ \quad\mp2\mathrm{Re}(iG^*\exp^{i(\Omega t-\mathbf{kr})}\rho),\\ \left(\frac{\partial}{\partial t}+(\mathbf{v}\nabla_\mathbf{r})+\Gamma\right)\rho=\nu\hat{\mathscr{V}}\rho-iG\exp^{-i(\Omega t-\mathbf{kr})}(\rho_m-\rho_n),\\ \hat{\mathscr{V}}=\frac{v_T^2}{2}\frac{\partial}{\partial v_\alpha}\Phi_{\alpha\beta}(\mathbf{v})\left(\frac{\partial}{\partial v_\beta}+\frac{2v_\beta}{v_T^2}\right),\end{cases} \tag{3}$$

where the "minus/plus" corresponds to the upper/lower level; ρ_{ij} is the particle density matrix ($\rho_j\equiv\rho_{jj}$ and $\rho\equiv\rho_{mn}$); Γ_m, Γ_n, and Γ are the relaxation constants of the states $|m\rangle$, $|n\rangle$, and the off−diagonal elements of the ρ_{ij} matrix; $G=\mathbf{E}_0\langle m|\hat{\mathbf{d}}|n\rangle/2\hbar$, with $\hat{\mathbf{d}}$ the dipole moment operator; $Q_mW(\mathbf{v})$ and $Q_nW(\mathbf{v})$ are the excitation functions of the states $|m\rangle$ and $|n\rangle$; $\hat{\mathscr{V}}$ describes elastic scattering; $\Phi_{\alpha\beta}(\mathbf{v})$ is the diffusion tensor; and ν is the transport collision rate, or the reciprocal of the time it takes a particle to change its velocity by a quantity on the order of the velocity proper.

When two particles are involved in the scattering process, there is only one preferred direction, that of their relative velocity. Hence the diffusion tensor $\Phi_{\alpha\beta}(\mathbf{v})$ has a single preferred axis, the direction of the velocity vector $\mathbf{v}$, and can be written as

$$\Phi_{\alpha\beta}(\mathbf{v})=\Phi_\parallel(v)\frac{v_\alpha v_\beta}{v^2}+\Phi_\perp\left(\delta_{\alpha\beta}-\frac{v_\alpha v_\beta}{v^2}\right), \tag{4}$$

where the longitudinal and transverse diffusion coefficients $\Phi_\parallel(v)$ and $\Phi_\perp(v)$ depend only on the absolute value of velocity. Here and in what follows velocity is measured in units of v_T and frequency in units of kv_T.

In this paper we obtain the shapes of the Bennett dip and nonlinear resonance without making any assumptions about the velocity dependence of the diffusion tensor. However, the analysis of the expressions and numerical calculations are done for ions in an equilibrium plasma, with

$$\Phi_\parallel(v)=3\int_0^1 d\lambda\lambda^2\,e^{-\lambda^2v^2},$$

$$\Phi_\perp=\frac{3}{2}\int_0^1 d\lambda(1-\lambda^2)\,e^{-\lambda^2v^2}. \tag{5}$$

Both $\Phi_\parallel(v)$ and $\Phi_\perp(v)$ decrease as functions of velocity. This is caused by the decrease in the Coulomb scattering cross section as the energy of the colliding particles grows.

3. THE VELOCITY DISTRIBUTION OF ρ_j

We solve the system of equations (3) by employing a perturbation expansion in the parameter $|G|^2$. The velocity distribution density $\rho_j(\mathbf{v})$ for particles that do not interact with the field and are in the state $|j\rangle$ has the form of the Maxwellian distribution $\rho_j^{(0)}(\mathbf{v})=Q_jW(\mathbf{v})/\Gamma_j$. The first-order field correction to $\rho_j(\mathbf{v})$ can be written as

$$\rho_j^{(1)}(\mathbf{v})=\mp2|G|^2N_{mn}^{(0)}\,\mathrm{Re}\int_0^\infty dt_2\exp(-\Gamma_jt_2+\nu\hat{\mathscr{B}}t_2)$$

$$\times\int_0^\infty dt_1\exp\{\hat{\mathscr{A}}t_1+\nu\hat{\mathscr{B}}t_1\},$$

$$\hat{\mathscr{A}}=-(\Gamma-i(\Omega-\mathbf{kv})),\qquad \hat{\mathscr{B}}=\frac{1}{2}\left(\frac{\partial}{\partial v_\alpha}-2v_\alpha\right)\Phi_{\alpha\beta}(\mathbf{v})\frac{\partial}{\partial v_\beta},$$

$$\hat{\mathscr{V}}f(\mathbf{v})W(\mathbf{v})=W(\mathbf{v})\hat{\mathscr{B}}f(\mathbf{v}),$$

where $N_{mn}^{(0)}=\rho_m^{(0)}(v)-\rho_n^{(0)}(v)$.

For $\nu\ll\Gamma_j\ll1$, which is typical of an argon laser plasma, we can explicitly calculate the operator exponents in (6) by employing the commutation relations between the operators $\hat{\mathscr{A}}$ and $\hat{\mathscr{B}}$:

$$\hat{\mathscr{B}}_1=[\hat{\mathscr{B}},\hat{\mathscr{A}}]$$

$$=i\left(v_z\Phi_\parallel(v)-\Phi_{\alpha z}(\mathbf{v})\frac{\partial}{\partial v_\alpha}-\frac{1}{2}\frac{\partial}{\partial v_\alpha}\Phi_{\alpha z}(\mathbf{v})\right),$$

$$\hat{\mathscr{B}}_2=[\hat{\mathscr{B}}_1,\hat{\mathscr{A}}]=-\Phi_{zz}(\mathbf{v}),$$

$$\hat{\mathscr{B}}_3=[\hat{\mathscr{B}}_2,\hat{\mathscr{A}}]=0,$$

$$\exp(\hat{\mathscr{A}}t+\nu\hat{\mathscr{B}}t)=\exp(\hat{\mathscr{A}}t)\exp\left(\frac{v}{6}\hat{\mathscr{B}}_2t^3+O(\nu t^2,\nu^2t^5)\right),$$

$$\exp(\nu\hat{\mathscr{B}}t_2)\exp(\hat{\mathscr{A}}t_1)$$

$$=\exp(\hat{\mathscr{A}}t_1)\exp\left(\nu\hat{\mathscr{B}}t_2+\nu\hat{\mathscr{B}}_1t_1t_2+\frac{\nu}{2}\hat{\mathscr{B}}_2t_1^2t_2\right).$$

Here the z axis is directed along $\mathbf{k}$. As a result, in the limit of small values of ν we have

$$\rho_j^{(1)}(\mathbf{v}) = \mp 2|G|^2 N_{mn}^{(0)} \operatorname{Re} \int_0^\infty dt_2 \int_0^\infty dt_1 \times \exp(-\Gamma_j t_2 + \hat{\mathscr{A}} t_1) \mathscr{R}(t_1, t_2, \mathbf{v}), \quad (7)$$

$$\mathscr{R}(t_1,t_2,\mathbf{v}) = \exp\left(\nu \hat{\mathscr{B}} t_2 + \nu \mathscr{B}_1 t_1 t_2 + \frac{\nu}{2} \hat{\mathscr{B}}_2 t_1^2 t_2 \right) \times \exp\left(\frac{\nu}{6} \hat{\mathscr{B}}_2 t_1^3 + O(\nu t_1^2, \nu^2 t_1^5) \right).$$

The integrand on the right−hand side of (7) is largest in the region $0 < t_2 < \Gamma_j^{-1}$, $0 < t_1 < \min(\Gamma^{-1}, \Gamma_j^{1/2}\nu^{-1/2}, \nu^{-1/3})$. Hence the terms $\nu \hat{\mathscr{B}} t_2$ and $O(\nu t_1^2, \nu^2 t_1^5)$ are characterized by a smallness of order $\nu \Gamma_j^{-1}$ and $\min(\nu^2\Gamma^{-5}, \Gamma_j^{5/2}\nu^{-1/2}, \nu^{1/3})$, respectively. Ignoring these terms, we can calculate the integral with respect to t_2 in (7):

$$\rho_j^{(1)}(\mathbf{v}) \simeq \mp 2|G|^2 N_{mn}^{(0)} \operatorname{Re} \int_0^\infty dt_1 \times \frac{\exp(-\Gamma t_1 + i(\Omega - v_z)t_1 - (\nu/6)\Phi_{zz}(\mathbf{v}) t_1^3)}{\Gamma_j + (\nu/2)\Phi_{zz}(\mathbf{v}) t_1^2 - i\nu \Phi_1(\mathbf{v}) t_1}, \quad (8)$$

where $\Phi_1(\mathbf{v}) = v_z \Phi_\parallel(v) - \frac{1}{2}(\partial \Phi_{\alpha z}(\mathbf{v})/\partial v_\alpha)$.

To establish the dependence of $\rho_j^{(1)}$ on v_z we must average $\rho_j^{(1)}(\mathbf{v})$ over the velocities $\mathbf{v}_\perp$, which are perpendicular to the z axis. If $\Phi_\parallel(v) = \Phi_\perp(v) \equiv 1$ (a constant diffusion coefficient), after averaging over $\mathbf{v}_\perp$ we get

$$\rho_j^{(1)}(v_z) \simeq \mp \frac{2|G|^2}{\sqrt{\pi}} \left(\frac{Q_m}{\Gamma_m} - \frac{Q_n}{\Gamma_n} \right) \exp^{-v_z^2} \times \operatorname{Re} \int_0^\infty dt_1 \frac{\exp(-\Gamma t_1 + i(\Omega - v_z)t_1 - (\nu/6)t_1^3)}{\Gamma_j + (\nu/2)t_1^2 - i\nu v_z t_1}. \quad (9)$$

Generally, the averaging can be done approximately by employing the renormalization method.[9] Here the method amounts to representing $\Phi_{zz}(\mathbf{v})$ and $\Phi_1(\mathbf{v})$ by the sums $\bar{\Phi}_{zz}(v_z, t_1) + \delta\Phi_{zz}$ and $\bar{\Phi}_1(v_z, t_1) + \delta\Phi_1$, respectively. The functions $\bar{\Phi}_{zz}(v_z, t_1)$ and $\bar{\Phi}_1(v_z, t_1)$ are chosen so that the first-order correction in $\delta\Phi_{zz}$ and $\delta\Phi_1$ to the integrand (the terms linear in $\delta\Phi_{zz}$ and $\delta\Phi_1$) vanish when integration over $\mathbf{v}_\perp$ is completed. Consequently, we arrive at the following expressions for $\bar{\Phi}_{zz}(v_z, t_1)$ and $\bar{\Phi}_1(v_z, t_1)$:

$$\bar{\Phi}_{zz}(v_z, t_1) = \langle \Phi_{zz}(\mathbf{v}) \rangle_\perp, \quad \bar{\Phi}_1(v_z, t_1) = \langle \Phi_1(\mathbf{v}) \rangle_\perp, \quad (10)$$

$$\langle f(\mathbf{v}) \rangle_\perp = \frac{1}{\pi} \int d^3\mathbf{v}_\perp f(\mathbf{v}) \exp^{-\mathbf{v}_\perp^2}.$$

For $\rho_j^{(1)}(v_z)$ we get

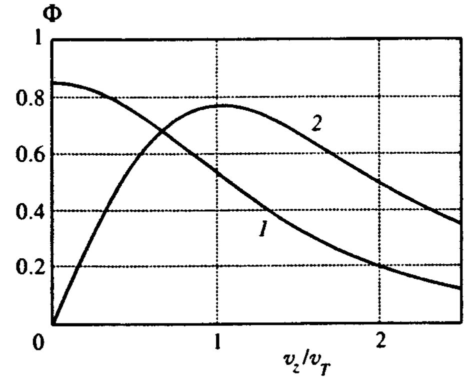

FIG. 1. The functions $\bar{\Phi}_{zz}(v_z)$ (curve *1*) and $\bar{\Phi}_1(v_z)$ (curve *2*) of v_z/v_T in an equilibrium plasma.

$$\rho_j^{(1)}(v_z) \simeq \mp \frac{2|G|^2}{\sqrt{\pi}} \left(\frac{Q_m}{\Gamma_m} - \frac{Q_n}{\Gamma_n} \right) \exp^{-v_z^2} \operatorname{Re} \int_0^\infty dt_1 \times \frac{\exp(-\Gamma t_1 + i(\Omega - v_z)t_1 - (\nu/6)\bar{\Phi}_{zz}(v_z) t_1^3)}{\Gamma_j + (\nu/2)\bar{\Phi}_{zz}(v_z) t_1^2 - i\nu \bar{\Phi}_1(v_z) t_1}. \quad (11)$$

Integrating (5), for the ions in the plasma we get

$$\bar{\Phi}_{zz}(v_z) = 6\pi \int_0^1 d\lambda \frac{\lambda^2}{(1+\lambda^2)^2} \exp^{-\lambda^2 v_z^2},$$

$$\bar{\Phi}_1(v_z) = 6\pi v_z \int_0^1 d\lambda \frac{\lambda^2}{1+\lambda^2} \exp^{-\lambda^2 v_z^2}. \quad (12)$$

Figure 1 depicts the decrease in $\bar{\Phi}_{zz}(v_z)$ and $\bar{\Phi}_1(v_z)$ with increasing v_z. Such behavior is caused by the decrease in the collision rate as the particle velocity grows. The difference between (11) and (9) is that ν is replaced with a function of v_z, which allows taking into account the velocity dependence of the diffusion tensor. In numerical calculations the deviation of the correction term $\rho_j^{(1)}(v_z)$ calculated via (11) and via (8) amounted to no more than 1%, which illustrates the high accuracy of the renormalization method.

The particles that interact with the field most strongly are those for which the detuning Ω is zero in their reference frame. These particles have a longitudinal velocity $v_z = \Omega$. Hence the function $\rho_j^{(1)}(v_z)$ has a peak (or dip) in the vicinity of $v_z = \Omega$ (Fig. 2). As the parameter ν/Γ_j grows, the distribution $\rho_j^{(1)}(v_z)$ broadens, since the interaction time $t_1^* \sim \sqrt{\Gamma_j/\nu}$ decreases because of diffusive loss of particles from the resonance state.

In a rough approximation the profile of $\rho_j^{(1)}(v_z)$ is the convolution of a Lorentzian profile of width Γ and a profile of the form $\exp\{-|v_z|\sqrt{\nu \bar{\Phi}_{zz}(v_z)/2\Gamma_j}\}$. For $\Gamma^2 \ll \nu \Gamma_h^{-1}$, the width of the profile of $\rho_j^{(1)}(v_z)$ is proportional to $\sqrt{\nu \bar{\Phi}_{zz}/\Gamma_j}$.

As Ω grows, the scattering rate for particles in resonance, which is proportional to the longitudinal component $\bar{\Phi}_{zz}(\Omega)$, decreases, as a result of which the distribution $\rho_j^{(1)}(v_z)$ may narrow (see Fig. 2). In the model with a constant diffusion tensor, the profile of Bennett's peak is a contour centered at $v_z = \Omega$ and with a Ω-independent shape, multiplied by the Maxwellian particle distribution function $\exp\{-v_z^2\}$. Hence Bennett's peak is asymmetric and its width increases with Ω. The effect of an increasing peak width at half-maximum becomes more pronounced as Γ grows, since the width of the peak approaches the Doppler value.

4. THE SPECTRUM OF THE PROBE FIELD

Now let us introduce another electromagnetic wave, the probe wave

$$\mathbf{E}_\mu(t,\mathbf{r}) = \tfrac{1}{2}(\mathbf{E}_{\mu 0}\exp\{-i(\omega_\mu \mathbf{t} - \mathbf{k}_\mu \mathbf{r})\} + \mathbf{E}^*_{\mu 0}\exp\{i(\omega_\mu \mathbf{t} - \mathbf{k}\mathbf{r})\}). \tag{13}$$

The condition that the probe field be weak means that $|G_\mu| \ll |G|$, where $|G_\mu| = \mathbf{E}_{\mu 0}[*]\langle m|\hat{\mathbf{d}}|n\rangle/2\hbar$. We assume that the probe wave also interacts resonantly with the two-level system, i.e., $\Omega_\mu = \omega_m - \omega_{mn} \ll \omega_\mu$. The wave vector $\mathbf{k}$ is assumed to be directed along the z axis (in the positive or negative direction). Since

$$\omega = |\mathbf{k}|c, \quad \omega_\mu = |\mathbf{k}_\mu|c, \quad |\mathbf{k}| - |\mathbf{k}_\mu| = \frac{\Omega - \Omega_\mu}{c} \sim \frac{kv_T}{c}$$

(at $\Omega, \Omega_\mu \sim kv_T$), we everywhere neglect the difference between the wave numbers. In this approximation, we have $\mathbf{k}_\mu = \kappa_\mu \mathbf{k}$, with $\kappa_\mu = \pm 1$.

The expression for the power absorbed from the probe wave is

$$\mathscr{P}(\Omega,\Omega_\mu) = -2\hbar\omega_{mn}\int d\mathbf{v}\,\mathrm{Re}(iG^*_\mu \exp^{i(\Omega_\mu t - \mathbf{k}_\mu \mathbf{r})}\rho). \tag{14}$$

To find ρ we must modify the system of equations (3) by introducing the interaction with the probe field:

$$\begin{cases} \left(\dfrac{\partial}{\partial t} + (\mathbf{v}\nabla_\mathbf{r}) + \Gamma_j\right)\rho_j = Q_j W(\mathbf{v}) + \nu\hat{\mathscr{V}}\rho_j \\ \quad \mp 2\,\mathrm{Re}(i(G^*\exp^{i(\Omega t - z)} + G^*_\mu \exp^{i(\Omega_\mu t - \kappa_\mu z)})\rho), \\ \left(\dfrac{\partial}{\partial t} + (\mathbf{v}\nabla_\mathbf{r}) + \Gamma\right)\rho = \nu\hat{\mathscr{V}}\rho - i \\ \quad \times(G\exp^{-i(\Omega t - z)} + G_\mu \exp^{-i(\Omega_\mu t - \kappa_\mu z)})(\rho_m - \rho_n). \end{cases} \tag{15}$$

We now write $\mathscr{P}(\Omega,\Omega_\mu)$ in the form of a series expansion in powers of intensity:

$$\mathscr{P}(\Omega,\Omega_\mu) = \sum_{k=1}^{\infty}\sum_{l=0}^{\infty}|G_\mu|^{2k}|G|^{2l}\mathscr{P}^{(k)(l)}(\Omega,\Omega_\mu).$$

The indices k and l correspond to the order of the perturbation in $|G|^2$ and $|G_\mu|^2$, respectively. At $k=1$ and $l=0$ the quantity $|G_\mu|^2\mathscr{P}^{(1)(0)}(\Omega,\Omega_\mu)$ is the absorbed power in the approximation in which the effect of the strong wave and the transitions induced by the probe wave are ignored. The correction to the absorbed power due to transitions caused by the probe wave is proportional to $|G_\mu|^4$ and is ignored here.

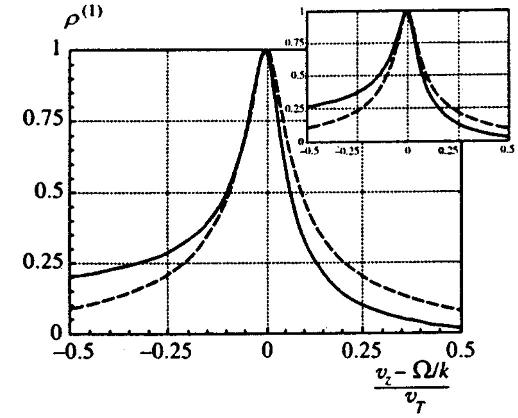

FIG. 2. The functions $\rho_j^{(1)}(v_z)$ at $\Omega = 0$ (the dashed curves) and $\Omega = kv_T$ (the solid curve), with $\Gamma = 10^{-2}kv_T$, $\Gamma_j = 10^{-3}kv_T$, and $\nu/\Gamma_j = 10^{-2}$. For a constant diffusion tensor (the inset) the width of the peak increases with Ω.

For $\nu \ll \Gamma_j \ll 1$ the change in the shape of $\mathscr{P}^{(1)(0)}(\Omega_\mu)$ brought on by Coulomb scattering is insignificant, with the result that we must examine $\mathscr{P}^{(1)(1)}(\Omega,\Omega_\mu)$, whose expression can be written as

$$\mathscr{P}^{(1)(1)}(\Omega,\Omega_\mu) = 2\hbar\omega_{mn}\,\mathrm{Re}\sum_{j=m,n}\sum_{i=1}^{4}\mathscr{P}_{i,j}(\Omega,\Omega_\mu), \tag{16}$$

where the $\mathscr{P}_{i,j}(\Omega,\Omega_\mu)$ are the different contributions to $\mathscr{P}^{(1)(1)}(\Omega,\Omega_\mu)$ corresponding to the different interaction processes:

$$\mathscr{P}_{i,j}(\Omega,\Omega_\mu) = \int d\mathbf{v}N^{(0)}_{mn}\int_0^\infty dt_1\int_0^\infty dt_2\int_0^\infty dt_3 \times \exp(\hat{\mathscr{A}}_\mu t_3 + \nu\hat{\mathscr{B}}t_3)\exp(\alpha_{i,j}t_2\}\exp\{\beta_i t_1),$$

$$\alpha_{1,j} = \alpha_{2,j} = -\Gamma_j + \nu\hat{\mathscr{B}},$$

$$\alpha_{3,j} = \alpha_{4,j} = -\Gamma_j - i(\Omega_\mu - \Omega) + i(\kappa_\mu - 1)v_z + \nu\hat{\mathscr{B}},$$

$$\beta_1^* = \beta_2 = \beta_4 = -\Gamma - i(\Omega - v_z) + \nu\hat{\mathscr{B}},$$

$$\beta_3 = -\Gamma + i(\Omega_\mu - \kappa_\mu v_z) + \nu\hat{\mathscr{B}}.$$

The quantities $\mathscr{P}_{1,j}$ and $\mathscr{P}_{2,j}$ correspond to cascade processes, i.e., a sequence of interactions with the photons of the strong and probe fields, and $\mathscr{P}_{3,j}$ and $\mathscr{P}_{4,j}$ to two-photon processes,[4] i.e., a simultaneous interaction with two photons of the electromagnetic field and transitions through a virtual level. The quantities $\mathscr{P}_{1,j}$ and $\mathscr{P}_{2,j}$ are caused by changes in the level populations brought about by the strong field (the saturation effect), while $\mathscr{P}_{3,j}$ and $\mathscr{P}_{4,j}$ describe the nonlinear interference effect and field splitting.

When $\kappa_\mu \simeq 1$ holds, the term with $(\kappa_\mu - 1)v_z$ can be neglected in comparison to Γ_j. But if we have $\kappa_\mu \simeq -1$, both $\mathscr{P}_{3,j}(\Omega,\Omega_\mu)$ and $\mathscr{P}_{4,j}(\Omega,\Omega_\mu)$ rapidly oscillate in v_z and after integration with respect to v_z yield a negligible contribution to $\mathscr{P}^{(1)(1)}(\Omega,\Omega_\mu)$ in comparison to $\mathscr{P}_{1,j}(\Omega,\Omega_\mu)$ and $\mathscr{P}_{2,j}(\Omega,\Omega_\mu)$. We analyze the expressions for $\mathscr{P}_{i,j}(\Omega,\Omega_\mu)$

for the case of codirectional waves we have, $\kappa_\mu=1$, since for calculating $\mathscr{P}^{(1)(1)}(\Omega,\Omega_\mu)$ in the case where $\kappa_\mu=1$ holds one can use the expression

$$\mathscr{P}^{(1)(1)}(\Omega,\Omega_\mu)\Big|_{\kappa_\mu=-1} \simeq 2\hbar\omega_{mn}\,\mathrm{Re}\sum_{j=m,n}\sum_{i=1}^{2}\mathscr{P}_{i,j}(\Omega,-\Omega_\mu)\Big|_{\kappa_\mu=1}. \qquad (17)$$

We introduce a new variable $\tau=t_3+t_1$ in $\mathscr{P}_{1,j}(\Omega,\Omega_\mu)$ and $\mathscr{P}_{3,j}(\Omega,\Omega_\mu)$ and another new variable $\tau=t_3-t_1$ in $\mathscr{P}_{2,j}(\Omega,\Omega_\mu)$ and $\mathscr{P}_{4,j}(\Omega,\Omega_\mu)$. This removes the terms of the form $v_z t_1$ in the exponent. In the first case we have to integrate with respect to τ from t_1 to ∞ and in the second from $-t_1$ to ∞. Hence the contributions of $\mathscr{P}_{1,j}(\Omega,\Omega_\mu)$ and $\mathscr{P}_{3,j}(\Omega,\Omega_\mu)$ are small compared to those of $\mathscr{P}_{2,j}(\Omega,\Omega_\mu)$ and $\mathscr{P}_{4,j}(\Omega,\Omega_\mu)$, respectively (this can be verified by analyzing the expressions obtained for $\mathscr{P}_{2,j}$ and $\mathscr{P}_{4,j}$; the above assumption has also proved to be true in numerical experiments).

The expressions for $\mathscr{P}_{i,j}(\Omega,\Omega_\mu)$ are integrals with respect to t_1, t_2, and t_3 that acquire their values in the region $0<t_3<\Gamma^{-1}$, $0<t_2<\Gamma_j^{-1}$, $0<t_1<\min(\Gamma^{-1}, \Gamma_j^{1/2}\nu^{-1/2}, \nu^{-1/3})$. Assuming that $\nu\ll\Gamma_j\ll 1$ and using commutation relations similar to those in Sec. 3 between the operators $\alpha_{i,j}$, β_i, and $\hat{\mathscr{B}}$, we get

$$\mathscr{P}_{2,j}(\Omega,\Omega_\mu)\simeq\int d\mathbf{v}N_{mn}^{(0)}\int_0^\infty dt_1 \times\frac{\exp\{-2\Gamma t_1-i(\Omega-\Omega_\mu)t_1-(\nu/3)\Phi_{zz}(\mathbf{v})t_1^3\}}{\Gamma_j+(\nu/2)\Phi_{zz}(\mathbf{v})t_1^2} \times\int_{-t_1}^{\infty}d\tau\,\exp(-\Gamma\tau+i(\Omega_\mu-v_z)\tau), \qquad (18)$$

$$\mathscr{P}_{4,j}(\Omega,\Omega_\mu)\simeq\int d\mathbf{v}N_{mn}^{(0)}\int_0^\infty dt_1 \times\frac{\exp\{-2\Gamma t_1-i(\Omega-\Omega_\mu)t_1-(\nu/3)\Phi_{zz}(\mathbf{v})t_1^3\}}{\Gamma_j+i(\Omega-\Omega_\mu)+(\nu/2)\Phi_{zz}(\mathbf{v})t_1^2} \times\int_{-t_1}^{\infty}d\tau\,\exp(-\Gamma\tau+i(\Omega_\mu-v_z)\tau). \qquad (19)$$

The terms $(\nu/6)\Phi_{zz}(\mathbf{n})\tau^3$ in the exponent have been discarded because they make a negligible contribution after integration with respect to v_z. We write $\Phi_{zz}(\mathbf{v})$ as the sum $\bar{\Phi}_{zz}(t_1,\Omega,\Omega_\mu)+\delta\Phi_{zz}$, where the first-order correction in $\delta\Phi_{zz}$ to the integrand (the terms linear in $\delta\Phi_{zz}$) vanish in the process of integration with respect to $\mathbf{v}$. The term $\bar{\Phi}_{zz}(t_1,\Omega,\Omega_\mu)$ must satisfy the equation

$$\int dv_z\,\exp^{-v_z^2}\int_{-t_1}^{\infty}d\tau\,\exp(-\Gamma\tau+i(\Omega_\mu-v_z)\tau) \times(\langle\Phi_{zz}(\mathbf{v})\rangle_\perp-\bar{\Phi}_{zz}(t_1,\Omega,\Omega_\mu))=0. \qquad (20)$$

After integration with respect to τ a narrow profile (of width Γ) emerges, centered at $v_z=\Omega_\mu$. We can therefore assume that

$$\bar{\Phi}_{zz}(t_1,\Omega,\Omega_\mu)\simeq\langle\Phi_{zz}(\mathbf{v})\rangle_\perp\Big|_{v_z=\Omega_\mu}=\bar{\Phi}_{zz}(\Omega_\mu).$$

Replacing $\Phi_{zz}(\mathbf{v})$ by its average value $\bar{\Phi}_{zz}(\Omega_\mu)$ and integrating Eqs. (18) and (19) first with respect to v_z and then with respect to τ, we arrive at the following expression for the nonlinear correction to the power absorbed from the probe field for the case $\kappa_\mu=1$:

$$\mathscr{P}^{(1)(1)}(\Omega,\Omega_\mu)\simeq 4\sqrt{\pi}\hbar\omega_{mn}\left(\frac{Q_m}{\Gamma_m}-\frac{Q_n}{\Gamma_n}\right)\exp^{-\Omega_\mu^2} \times\mathrm{Re}\sum_{j=m,n}\left[\int_0^\infty dt_1\frac{\exp(-2\Gamma t_1-i(\Omega-\Omega_\mu)t_1-(\nu/3)\bar{\Phi}_{zz}(\Omega_\mu)t_1^3)}{\Gamma_j+(\nu/2)\bar{\Phi}_{zz}(\Omega_\mu)t_1^2} +\int_0^\infty dt_1\frac{\exp(-2\Gamma t_1-i(\Omega-\Omega_\mu)t_1-(\nu/3)\bar{\Phi}_{zz}(\Omega_\mu)t_1^3)}{\Gamma_j+i(\Omega-\Omega_\mu)+(\nu/2)\bar{\Phi}_{zz}(\Omega_\mu)t_1^2}\right]. \qquad (21)$$

When $\kappa_\mu=-1$ holds, we must replace Ω_μ by $-\Omega_\mu$ and discard the second term in the square brackets.

The first term in the square brackets is the population correction to the absorbed power. If we replace Γ by $\Gamma/2$ and Ω_μ by v_z, it coincides, within a factor, with Eq. (11) for the nonlinear correction to the population difference $\rho_m^{(1)}(v_z)-\rho_n^{(1)}(v_z)$. This means that at $\kappa_\mu=-1$ the contour for $\mathscr{P}^{(1)(1)}(\Omega,\Omega_\mu)$ has the same shape as the contour for $\rho_m^{(1)}\times(v_z)-\rho_n^{(1)}(v_z)$ for a double value of Γ. Like the Bennett dip, at $\kappa_\mu=-1$ the probe field spectrum $\mathscr{P}^{(1)(1)}(\Omega,\Omega_\mu)$ as a function of Ω_μ may narrow as Ω grows (Fig. 3). The effect of spectrum narrowing is greatest when the diffusion width is much smaller than the Doppler width but much greater than the homogeneous width Γ, or $\Gamma/kv_T\ll\sqrt{\nu/\Gamma_j}\ll 1$.

When $\kappa_\mu=1$ holds, we must allow for the second term, which corresponds to two-photon processes. In the region

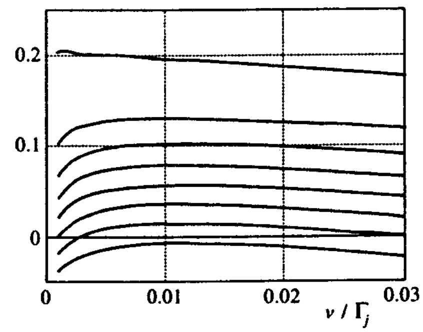

FIG. 3. The ν/Γ_j-dependence of the relative narrowing of the absorption spectrum $\mathscr{P}^{(1)(1)}(\Omega,\Omega_\mu)$ as Ω varies from 0 to kv_T (for $\kappa_\mu=-1$), obtained as a result of numerical calculations by Eq. (21). The different curves correspond to different values of Γ, from $\Gamma=0$ (the upper curve) to $\Gamma=7\times10^{-2}kv_T$, with a growth rate of $10^{-2}kv_T$.

$|\Omega_\mu-\Omega|\ll\nu^{1/3}$ with $\Gamma^2\Gamma_j\ll\nu$, the shape of the peak is proportional to the square root of the Lorentzian, just as it is in the model with a constant diffusion coefficient:[10]

$$\mathrm{Re}\,\mathscr{P}_{4,j}(\Omega,\Omega_\mu)\propto\mathrm{Re}\times e^{-\Omega_\mu^2}\sum_{j=m,n}\left[\frac{\nu}{2}\bar{\Phi}_{zz}(\Omega_\mu)\times(\Gamma_j+i(\Omega_\mu-\Omega))\right]^{-1/2}. \quad (22)$$

The width of the peak is independent of the collision rate ν and, hence, of Ω, while the height of the peak is proportional to $\nu^{1/2}\bar{\Phi}_{zz}^{1/2}(\Omega_\mu)$ and coincides with that of the "population" peak $\mathscr{P}_{2,j}(\Omega,\Omega_\mu)$. In the region $|\Omega_\mu-\Omega|\gg\nu^{1/3}$ and to leading order in $\nu(\Omega_\mu-\Omega)^{-3}$, collisions have no effect on the lineshape, but the integral of $\mathscr{P}_{4,j}(\Omega,\Omega_\mu)$ with respect to Ω_μ vanishes, as it does in the absence of collisions (Fig. 4).

5. CONCLUSION

Summing up, we may say that in the case of Coulomb scattering of particles with $\Gamma\ll kv_T$, the probe field spectrum narrows as the frequency of the strong field departs from the center of the line. The physical reason for this is that the spectrum width is determined by the homogeneous width $\Delta\omega_{\mathrm{hom}}=2\Gamma$ and by the diffusion width $\Delta\omega_{\mathrm{dif}}$, the latter depending on the collision rate: $\Delta\omega_{\mathrm{dif}}\sim kv_T\sqrt{\nu\tau_j}$, where τ_j is the ion lifetime in the state $|j\rangle$, $\tau_j=\Gamma_j^{-1}$. As we move away from resonance, the wave begins to interact with faster ions. The collision rate ν for fast ions is low, since the Rutherford cross section decreases as the energy grows. Thus, as the detuning from resonance increases, the diffusion width $\Delta\omega_{\mathrm{dif}}\propto\sqrt{\nu}$ decreases. It would be more correct to call this a decrease of diffusion width rather than spectrum narrowing. As $\Delta\omega_{\mathrm{hom}}\to0$ and the detuning varies from 0 to kv_T, the relative narrowing begins at $1-\sqrt{\bar{\Phi}_{zz}(v_T)/\bar{\Phi}_{zz}(0)}$ and reaches 20% (see Fig. 3), where $\bar{\Phi}_{zz}(v_z)=\langle\Phi_{zz}(\mathbf{v})\rangle_\perp$ is the zz-component of the diffusion tensor averaged over transverse velocities (see Fig. 1).

The effect may be masked by atomic collisions with a different velocity dependence of the cross section and by Stark broadening. In an argon-laser plasma with characteristic parameters (the plasma temperature is 1 eV and the ion number density is 1/100 of the atom number density), the Coulomb scattering is stronger than ion–atom scattering. For an electron number density $N_e\sim10^{14}\mathrm{cm}^{-3}$ and an electron temperature $T_e\simeq5$ eV, the Stark linewidth of the ion is of order $10^8\mathrm{s}^{-1}$, which is much smaller that the Doppler linewidth $kv_T\sim10^{10}\mathrm{s}^{-1}$. For parameter ratios characteristic of an argon-laser plasma, $\Gamma\sim10^{-2}kv_T$ and $\nu\Gamma_j^{-1}\sim10^{-2}$, the relative narrowing amounts to about 10%, i.e., the effect is experimentally observable.

Of particular interest is the direct measurement of the diffusion tensor components in the velocity space as functions of the particle velocity. Bowles *et al.*[11] described another way of measuring the diffusion tensor. A powerful short light pulse generates a Bennett structure, whose evolution is then studied. This method is possible only when the ion number density N_i is low (otherwise the structure spreads out too fast, and this would require high-speed registering devices; in Ref. 11 N_i did not exceed $5\times10^9\mathrm{cm}^{-3}$). Our method can be used at low and high ion number densities. It can be employed for measuring the averaged diffusion tensor $\bar{\Phi}_{zz}(v_z)$ if the width of the probe field spectrum is largely determined by diffusion. The narrowing effect can be used to find the fraction of Coulomb scattering in the total cross section.

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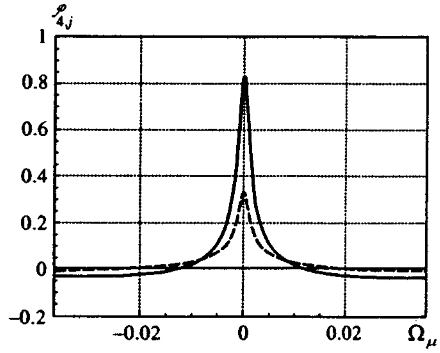

FIG. 4. The shape of the peak corresponding to two-photon processes for $\Gamma=10^{-2}kv_T$, $\Omega=0$, and $\Gamma_j=10^{-3}kv_T$ with $\nu=10^{-5}kv_T$ (the solid curve) and $\nu=10^{-4}kv_T$ (the dashed curve), in arbitrary units.

[1] P. R. Berman, Adv. At. Mol. Phys. **13**, 57 (1977).
[2] S. G. Rautian, Preprint No. 82, Institute of Automation and Electrometry, Siberian Branch of the Russian Academy of Sciences, Novosibirsk (1978).
[3] W. R. Bennett, Phys. Rev. **126**, 580 (1962).
[4] S. G. Rautian, G. I. Smirnov, and A. M. Shalagin, *Nonlinear Resonances in the Spectra of Atoms and Molecules*, Nauka, Novosibirsk (1979) [in Russian].
[5] S. A. Babin and D. A. Shapiro, Phys. Rep. **241**, 119 (1994).
[6] E. V. Podivilov, A. I. Chernykh, and D. A. Shapiro, Zh. Éksp. Teor. Fiz. **105**, 1214 (1994) [JETP **78**, 653 (1994)].

[7] S. G. Rautian, Usp. Fiz. Nauk **161**, 151 (1991). [Sov. Phys. Uspekhi **34**, 1008 (1991)].
[8] E. V. Podivilov, D. A. Shapiro, and M. G. Stepanov, Phys. Rev. Lett. **74**, 3979 (1995).
[9] A. H. Nayfeh, *Introduction to Perturbation Techniques*, Wiley, New York (1981).
[10] S. G. Rautian and D. A. Shapiro, Zh. Éksp. Teor. Fiz. **94**, No. 10, 110 (1988) [Sov. Phys. JETP **67**, 2018 (1988)].
[11] J. Bowles, R. McWilliams, and N. Rynn, Phys. Rev. Lett. **68**, 1144 (1992).

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