Diffusion component of the profile of a nonlinear resonance in a three-level system

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The formation by diffusion of a nonlinear resonance in a three-level system with a large Doppler broadening is studied. An asymptotic expansion of the work performed by a test field is derived in the limit of intense diffusion. The profile of the resonance is found to consist of three components: a narrow peak, a broad part of diffusion origin, and Lorentzian wings. It is not possible to separate the narrow and broad components by varying the polarization of the fields.

1. INTRODUCTION

The diffusive motion of an atom in velocity space is known to result in a broadening of nonlinear spectral resonances.[1] Spectroscopic manifestations of particle diffusion in the field of an intense light wave have recently been studied in detail in the particular case of two-level systems.[2,3] A topic of special interest is the diffusion caused by Coulomb ion-ion scattering through small angles,[4] because of a possible application of nonlinear spectroscopy in plasma diagnostics.

To determine the physical nature of the diffusive broadening we consider the distribution of the difference between the populations of two atomic levels in the projection of the velocity **v** onto the wave vector of the light wave, **k**. A resonant wave "burns out" a Bennett dip or peak width Γ/k (Γ is the homogeneous linewidth) against the background of a Maxwellian distribution. The diffusion in velocity space with a coefficient D tends to smooth over the nonequilibrium structure. This structure spreads out, and the particles move out of resonance with the wave. The typical change in the velocity of an atom over the lifetime Γ_j^{-1} in level j is $(D/\Gamma_j)^{1/2}$. Since we have $D=\nu\bar{v}^2/2$, where ν is the transport collision rate, and $\bar{v}$ is the average thermal velocity, the ratio of the diffusion width to the homogeneous width, $(\nu/\Gamma_j)^{1/2}k\bar{v}/\Gamma$, contains an "amplifying factor" $k\bar{v}/\Gamma \gg 1$ and can be large even under the condition $\nu/\Gamma_j \ll 1$. In addition to acting on the velocity distribution, the diffusion causes a frequency modulation of the dipole moment; the parameter $p=\nu(k\bar{v})^2/\Gamma^3 = k^2D/\Gamma^3$ serves as a measure of the role which it plays.

Several experiments have been carried out to measure the width of the Lamb dip in the frequency dependence of the output power of argon-ion lasers at various charged-particle densities.[3,5] The results of these measurements agree with the qualitative picture drawn here; the diffusion width of the dip due to ion-ion collisions has exceeded the homogeneous width by a factor of three to five.

The test-field method differs from the Lamb-dip method in that it allows one to observe ultranarrow resonances caused by two-photon transitions or, according to ideas corresponding to resonant conditions, caused by nonlinear interference effects stemming from a mixing of states by a strong field. In these processes, there may be a complete or nearly complete cancellation of the Doppler shifts of the strong and test waves, with the result that the resonances in the spectrum of the test field are particularly narrow. It might seem that such resonances should undergo a giant broadening due to velocity diffusion, but experiments by Lebedeva *et al.*[8] ($\lambda = 4880$ and 5145 Å, ArII) did not reveal this broadening. Furthermore, when the line used as the test line was changed, the width of the resonance changed. Such unexpected experimental results force us to acknowledge that we do not have a clear picture of the basic theory of the test-field method.

Our purpose in the present study was to calculate the diffusion lineshape of a nonlinear resonance in a three-level system with a large Doppler broadening. In Sec. 2 we present a system of kinetic equations for a density matrix. We find Green's functions for these equations. In Sec. 3 we derive equations for the lineshape of a narrow nonlinear resonance in a Raman-scattering arrangement. We derive an asymptotic expansion for the limiting case of intense diffusion. The results show that the lineshape of the resonance is made up of three parts: a narrow peak, with a profile which is the square root of a Lorentzian profile; a broad diffusion part; and Lorentzian wings. Section 4 incorporates the degeneracy of the states in angular-momentum projections. It is found that—in contrast with the case of a two-level system—the narrow and broad components cannot be separated by varying the polarizations of the saturating and test fields. In Sec. 5 we compare this new theory with experimental data.[8]

2. KINETIC EQUATION AND GREEN'S FUNCTIONS

We consider a gas of particles which are interacting resonantly with an external electromagnetic field. In the Wigner representation, which is convenient for a classical description of the translational motion of atoms, the equation for the density matrix $\rho(\alpha,\alpha',\mathbf{r},\mathbf{v},t)$ takes the form[1,9]

$$\left(\frac{\partial}{\partial t}+\mathbf{v}\nabla_{\mathbf{r}}\right)\rho(\alpha,\alpha';\mathbf{r},\mathbf{v},t)=R(\alpha,\alpha')+S(\alpha,\alpha')$$
$$-i\sum_{\alpha_1}[V(\alpha,\alpha_1;\mathbf{r},t)\rho(\alpha_1,\alpha';\mathbf{r},\mathbf{v},t)$$
$$-\rho(\alpha,\alpha_1;\mathbf{r},\mathbf{v},t)V(\alpha_1,\alpha';\mathbf{r},t)], \qquad (2.1)$$

where $\alpha = \alpha JM$ is the set of quantum numbers, J is the total angular momentum, and M is its projection. The matrix R describes a radiative relaxation, and S is a collision integral. The operator V is the Hamiltonian of the interaction of the atom with the electromagnetic field. In Eq. (2.1) we are ignoring the effect of the external field on the translational degrees of freedom of the atom.

We first consider a three-level system without degeneracy (we will write $\alpha = j$, the index of the energy level, as a subscript). We assume that an atom is interacting with the electromagnetic radiation in a Raman-scattering arrangement through a lower level n (Fig. 1a). The strong field and

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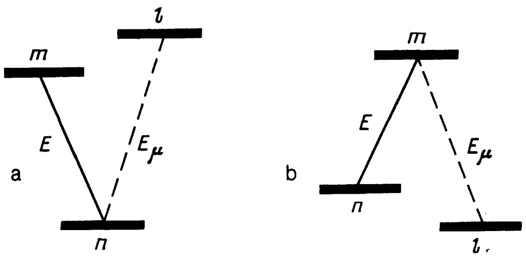

FIG. 1. Test-field spectroscopy. a—In an arrangement for Raman scattering through a lower level; b—through an upper level. a) V configuration; b) Λ configuration.

the test field are represented by the traveling waves

$$\mathbf{E}(\mathbf{r},t)=\mathbf{E}\exp(-i\omega t+i\mathbf{kr}),\quad \mathbf{E}_\mu(\mathbf{r},t)=\mathbf{E}_\mu\exp(-i\omega t+i\mathbf{k}_\mu\mathbf{r}),\tag{2.2}$$

which are in resonance with the m–n and l–n transitions. The matrix elements of the dipole interaction of the light with these transitions are

$$V_{mn}=-G\exp(-i\Omega t+i\mathbf{kr}),\quad G=-\mathbf{E}\mathbf{d}_{mn}/2\hbar,\ \Omega=\omega-\omega_{mn},\tag{2.3}$$

$$V_{ln}=-G_\mu\exp(-i\Omega_\mu t+i\mathbf{k}_\mu\mathbf{r}),\quad G_\mu=-\mathbf{E}_\mu\mathbf{d}_{ln}/2\hbar,\ \Omega_\mu=\omega_\mu-\omega_{ln},$$

where $\mathbf{d}_{jn}$ and ω_{jn} are the matrix elements of the dipole-moment operator and of the Bohr frequencies of the j–n transitions ($j=m,l$). The l–m transition is forbidden in the dipole approximation. We can seek the diagonal elements of the density matrix in a time-independent form, and the off-diagonal elements in an oscillatory form (for the forbidden transition at the Raman frequency $\varepsilon=\Omega_\mu-\Omega$, $\mathbf{q}=\mathbf{k}_\mu-\mathbf{k}$):

$$\rho_{jj}=r_{jj},\ \rho_{mn}=r_{mn}\exp(-i\Omega t+i\mathbf{kr}),\ \rho_{ln}=r_{ln}\exp(-i\Omega_\mu t+i\mathbf{k}_\mu\mathbf{r}),\ \rho_{ml}=r_{ml}\exp(i\varepsilon t-i\mathbf{qr}).\tag{2.4}$$

Substituting (2.4) into (2.1), we find the following steady-state system of equations for the amplitudes r_{ij}:

$$\begin{gathered}\Gamma_{mm}r_{mm}=q_m-2\operatorname{Re}(iG^*r_{mn})+S_{mm},\\ \Gamma_{ll}r_{ll}=q_l-2\operatorname{Re}(iG_\mu^*r_{ln})+S_{ll},\\ \Gamma_{nn}r_{nn}=q_n+2\operatorname{Re}(iG^*r_{mn}+iG_\mu^*r_{ln})+A_{mn}r_{mm}+A_{ln}r_{ll}+S_{nn},\\ [\Gamma_{mn}+i(\Omega-\mathbf{kv})]r_{mn}^*=iG^*(r_{mm}-r_{nn})+iG_\mu^*r_{ml}^*+S_{mn},\\ [\Gamma_{ln}-i(\Omega_\mu-\mathbf{k}_\mu\mathbf{v})]r_{ln}=-iG_\mu(r_{ll}-r_{nn})-iGr_{ml}^*+S_{ln},\\ [\Gamma_{ml}-i(\varepsilon-\mathbf{qv})]r_{ml}^*=-iG^*r_{ln}+iG_\mu r_{mn}^*+S_{ml},\end{gathered}\tag{2.5}$$

where Γ_{ij} are the relaxation constants of the levels and the polarizations, which incorporate both radiative processes and collisions which do not involve a change in velocity (the relaxation-constant model),[1)] A_{jn} are Einstein coefficients, and g_j are the level excitation functions, which we assume to be Maxwellian,

$$q_j(\mathbf{v})=\frac{Q}{(\pi^{1/2}\bar{v})^3}\exp\left(-\frac{\mathbf{v}^2}{\bar{v}^2}\right),\quad \bar{v}=\left(\frac{2T}{m}\right)^{1/2},\tag{2.6}$$

where T, and m are the temperature and mass of the particles. Taking the particles to be ions, and assuming that they scatter according to a Coulomb law, we assume that the rates ν and the kernels A of the collision integrals

$$S_{ij}(\mathbf{v})=-\nu(\mathbf{v})\rho_{ij}+\int A(\mathbf{v}|\mathbf{v}')\rho_{ij}(\mathbf{v}')\,d\mathbf{v}'\tag{2.7}$$

do not depend on the states; we assume that the collisions are elastic; and we assume that the kernels are of a difference form:

$$\int S_{ij}(\mathbf{v})\,d\mathbf{v}=0,\quad A(\mathbf{v}|\mathbf{v}')=A(\mathbf{v}-\mathbf{v}'),\quad \nu=\text{const}.\tag{2.8}$$

We are ignoring the slowing of the particles caused by the dynamic friction force.

We can find a solution of system of the integral equations (2.5) by perturbing in the field amplitudes G and G_μ. A perturbation theory is usually constructed with the help of Green's functions, which are in turn calculated by means of Fourier transforms. The calculations can be shortened slightly by changing the order of operations: by first taking Fourier transforms of Eqs. (2.5),

$$\Phi(\mathbf{v})=\int\frac{d\boldsymbol{\zeta}}{(2\pi)^3}e^{i\boldsymbol{\zeta}\mathbf{v}}\Phi(\boldsymbol{\zeta}),\quad \Phi(\boldsymbol{\zeta})=\int d\mathbf{v}e^{-i\boldsymbol{\zeta}\mathbf{v}}\Phi(\mathbf{v}),\tag{2.9}$$

and seeking the Green's functions in the ζ representation at once. Here Φ is any of the functions which figure in (2.5). The variable ζ, which is the conjugate of the velocity $\mathbf{v}$, has the meaning of a quantity which is proportional to a difference between coordinates, and transformation (2.9) is a transition from a Wigner representation to a coordinate representation of the density matrix. The integral equations (2.5) with a difference kernel reduce to differential equations after Fourier transforms are taken.

The equations for the diagonal (f_{jj}) and off-diagonal ($f_{ij}, i\neq j$) Green's functions are

$$\begin{gathered}(\Gamma_{jj}+\varphi)f_{jj}(\boldsymbol{\zeta}|\boldsymbol{\zeta}')=\delta(\boldsymbol{\zeta}-\boldsymbol{\zeta}'),\quad \varphi(\boldsymbol{\zeta})=\nu-A(\boldsymbol{\zeta}),\\ (\Gamma+\varphi-\mathbf{p}\nabla_{\boldsymbol{\zeta}})f_{ij}(\boldsymbol{\zeta}|\boldsymbol{\zeta}')=\delta(\boldsymbol{\zeta}-\boldsymbol{\zeta}'),\end{gathered}\tag{2.10}$$

and the complex parameter Γ and the vector $\mathbf{p}$ take on the following respective values for the functions f_{mn}, f_{ln}, and f_{mn}: $\Gamma_{mn}+i\Omega$, $-\mathbf{k}$; $\Gamma_{ln}-i\Omega_\mu$, $\mathbf{k}_\mu$; and $\Gamma_{ml}-i\varepsilon$, $\mathbf{q}$ [see Eqs. (2.5)]. The diagonal Green's function is simply

$$f_{jj}(\boldsymbol{\zeta}|\boldsymbol{\zeta}')=\frac{\delta(\boldsymbol{\zeta}-\boldsymbol{\zeta}')}{\Gamma_{jj}+\varphi(\boldsymbol{\zeta})},\tag{2.11}$$

while the off-diagonal Green's function takes its simplest form in a coordinate system whose z axis runs parallel to the vector $\mathbf{p}$:

$$f_{ij}(\boldsymbol{\zeta}|\boldsymbol{\zeta}')=\frac{\delta(\boldsymbol{\zeta}_\perp-\boldsymbol{\zeta}_\perp')\,\Theta(\sigma(\zeta_z-\zeta_z'))}{|\mathbf{p}|}\cdot\exp\left(-\sigma\int_{\zeta_z'}^{\zeta_z}\frac{\Gamma+\varphi(\boldsymbol{\zeta}_\perp,\zeta_z'')}{|\mathbf{p}|}d\zeta_z''\right),\tag{2.12}$$

$$\Theta(x)=\begin{cases}1, & x>0,\quad \zeta_z=\boldsymbol{\zeta}\mathbf{n},\qquad \sigma=\operatorname{sign}(\mathbf{pn})\\ 0, & x\leqslant 0,\quad \boldsymbol{\zeta}_\perp=\boldsymbol{\zeta}-\mathbf{n}\zeta_z\end{cases}.$$

Here $\mathbf{n}$ is a unit vector along the z axis. In the following sections of this paper we will be treating the case in which the vectors $\mathbf{k}$ and $\mathbf{k}_\mu$ are parallel, so that we can select a common z axis for all of the off-diagonal Green's functions.

The degeneracy of the states in the angular-momentum projections can be dealt with conveniently by expanding Eq. (2.1) in irreducible tensor operators (the $\varkappa q$ representation):

$$L_{ij}(\varkappa q)=\sum_{MM'}(-1)^{J'-M'}\langle JMJ'-M'|\varkappa q\rangle L(aJM,a'J'M'),$$
$$L(aJM,a'J'M')=\sum_{\varkappa q}(-1)^{J'-M'}\langle JMJ'-M'|\varkappa q\rangle L_{ij}(\varkappa q), \qquad (2.13)$$

where $L(aJM,a'J'M')$ is the matrix element of an arbitrary operator $\hat{L}$ in the basis of eigenfunctions of the angular-momentum operator $\hat{\mathbf{J}}$. If the collision integral is diagonal in $\varkappa q$ and does not depend on $\varkappa$ or q, the Green's functions are again diagonal:

$$f_{ij}(\varkappa q\zeta|\varkappa_1 q_1\zeta_1)=\delta_{\varkappa\varkappa_1}\delta_{qq_1}f_{ij\varkappa}(\zeta|\zeta_1). \qquad (2.14)$$

The diagonal (in $\varkappa$) functions $f_{ij\varkappa}$ satisfy the same equations as are satisfied by the functions f_{ij} in the model of nondegenerate states.

In the case of interest here, in which the waves are propagating in the same direction and have approximately equal frequencies ($\mathbf{k}_\mu\uparrow\uparrow\mathbf{k}$, $k_\mu\approx k$), we can ignore the gradient ∇_ζ in the equation for $f_{ml\varkappa}$; the equation then becomes an algebraic equation, like the equations for the diagonal elements $f_{jj\varkappa}$. Of the six we are left with only two differential equations, for $f_{mn\varkappa}$ and for $f_{ln\varkappa}$.

3. SHAPE OF THE NONLINEAR RESONANCE

Let us find the profile of a nonlinear resonance in a model of nondegenerate states. For this purpose we construct a perturbation theory in the strength of the optical field. In zeroth order, the matrix $r_{ij}(\zeta)$ is diagonal, and we find the following expressions for the populations:

$$r_{jj}(\zeta)=\frac{q_j(\zeta)}{\Gamma_{jj}+\varphi(\zeta)},\quad j=m,l,$$
$$r_{nn}(\zeta)=\frac{1}{\Gamma_{nn}+\varphi(\zeta)}\left[q_n(\zeta)+\sum_{j=l,m}\frac{q_j(\zeta)A_{jn}}{\Gamma_{jj}+\varphi(\zeta)}\right]. \qquad (3.1)$$

Thereafter, in odd orders in the field, polarizations appear on the allowed transitions r_{mn} and r_{ln}, while in even orders polarizations appear on the forbidden transition r_{ml}, along with corrections to the populations r_{jj} for the effect of saturation. The work performed by the field $\mathbf{E}_\mu$ on the l–n transition is given in the ζ representation by

$$P_\mu=2\hbar\omega_\mu\,\mathrm{Re}\left\{iG_\mu^*\int\delta(\zeta)r_{ln}(\zeta)d\zeta\right\}. \qquad (3.2)$$

The complete expression for the work performed by the field, (3.2), reduces, within the first nonvanishing corrections for the strong field, to the sum of four terms. There is a linear term proportional to the intensity of the test wave, $|G_\mu|^2$, and there are three nonlinear terms which contain the product of intensities $|G_\mu G|^2$:

$$P_\mu=P_\mu^{(0)}-\sum_{i=1}^{3}P_\mu^{(i)},$$
$$P_\mu^{(0)}=2\hbar\omega_\mu|G_\mu|^2\,\mathrm{Re}\langle\delta(\zeta_1)f_{ln}(\zeta_1|\zeta_2)N_{ln}(\zeta_2)\rangle,$$
$$P_\mu^{(1)}=2\hbar\omega_\mu|G_\mu G|^2\,\mathrm{Re}\langle\delta(\zeta_1)f_{ln}(\zeta_1|\zeta_2)f_{nn}(\zeta_2|\zeta_3)$$
$$\cdot[\delta(\zeta_3-\zeta_4)-A_{mn}f_{mm}(\zeta_3|\zeta_4)][f_{mn}(\zeta_4|\zeta_5)+\bar{f}_{mn}(\zeta_4|\zeta_5)]N_{mn}(\zeta_5)\rangle,$$
$$P_\mu^{(2)}=2\hbar\omega_\mu|G_\mu G|^2$$
$$\cdot\mathrm{Re}\langle\delta(\zeta_1)f_{ln}(\zeta_1|\zeta_2)f_{ml}(\zeta_2|\zeta_3)f_{mn}(\zeta_3|\zeta_4)N_{mn}(\zeta_4)\rangle,$$
$$P_\mu^{(3)}=2\hbar\omega_\mu|G_\mu G|^2$$
$$\cdot\mathrm{Re}\langle\delta(\zeta_1)f_{ln}(\zeta_1|\zeta_2)f_{ml}(\zeta_2|\zeta_3)f_{ln}(\zeta_3|\zeta_4)N_{ln}(\zeta_4)\rangle,$$
$$\bar{f}_{ij}(\zeta|\zeta';\,\Gamma,\,\mathbf{p})=f_{ij}(\zeta|\zeta';\,\Gamma^*,\,-\mathbf{p}). \qquad (3.3)$$

Here $N_{ij}=r_{ii}-r_{jj}$ is the difference between unperturbed populations (3.1), and the angle brackets indicate integration over all of the variables ζ_i inside the angle brackets.

The other terms of the nonlinear part of the work performed by the field correspond to three basic effects of nonlinear spectroscopy: $P_\mu^{(1)}$, a population term, corresponds to the effect of saturation; $P_\mu^{(2)}$ corresponds to nonlinear interference; and $P_\mu^{(3)}$ corresponds to field splitting. The last two effects are manifested only for copropagating waves, as in the collisionless case; the field splitting disappears at $k_\mu>k$.

Relations (3.3) hold for arbitrary kernels of the collision integrals. In the case of difference kernels, the Green's functions f_{jj} and f_{ij} are given by (2.11) and (2.12). For the Coulomb scattering in which we are interested here it is natural to adopt the diffusion approximation, in which we have $\varphi(\zeta)=D\zeta^2$, where D is a diffusion coefficient (we recall that we are not considering dynamic friction). We also assume that the Doppler width is significantly greater than the structure in which we are interested here: $k\bar{v}\gg|\Omega|$, Γ_{ij}, $(Dk^2/\Gamma_{ij})^{1/2}$. We can thus replace the Maxwellian distribution by a δ-function:

$$\exp[-(\bar{v}\zeta)^2/2]=(2\pi)^{3/2}\delta(\zeta)/\bar{v}^3.$$

As a result, expression (3.3) for the work performed by the field simplifies substantially. A linear absorption or amplification reduces to the familiar Doppler lineshape

$$P_\mu^{(0)}=2\hbar\omega_\mu\frac{\pi^{1/2}}{k\bar{v}}|G_\mu|^2N_{ln}(0)\exp(-\Omega_\mu^2/k_\mu^2\bar{v}^2). \qquad (3.4)$$

The nonlinear terms in the work performed by the field can be calculated by substituting the Green's functions (2.11) and (2.12) into expression (3.3). Here are the expressions found for the case of copropagating waves ($\mathbf{k}_\mu\uparrow\uparrow\mathbf{k}$), in which the resonance in the "bent" three-level system (Fig. 1) is found to be narrower[2)]:

$$P_\mu^{(1)}\propto\mathrm{Re}\int_0^\infty d\zeta\left(1-\frac{A_{mn}}{\Gamma_{mm}+D\zeta^2}\right)$$
$$\cdot\exp\left[-\left(\frac{\Gamma_{ln}-i\Omega_\mu}{k_\mu}+\frac{\Gamma_{mn}+i\Omega}{k}\right)\zeta-\frac{D}{3}\left(\frac{1}{k}+\frac{1}{k_\mu}\right)\zeta^3\right](\Gamma_{nn}+D\zeta^2)^{-1},$$
$$P_\mu^{(2)}\propto\mathrm{Re}\frac{1}{|k_\mu-k|}\int_0^\infty d\zeta''\int_0^{\zeta''}d\zeta'\exp\left[-\frac{\Gamma_{ln}-i\Omega_\mu}{k_\mu}\zeta'-\frac{\Gamma_{mn}+i\Omega}{k}\zeta''\right.$$
$$\left.-\frac{\Gamma_{ml}-i\varepsilon}{k_\mu-k}(\zeta''-\zeta')-\frac{D\zeta'^3}{3k_\mu}-\frac{D\zeta''^3}{3k}-\frac{D}{3}\frac{\zeta''^3-\zeta'^3}{k_\mu-k}\right],\quad k_\mu>k, \qquad (3.5)$$
$$P_\mu^{(3)}\propto\mathrm{Re}\frac{1}{k-k_\mu}\left\{\int_0^\infty d\zeta\exp\left[-\left(\frac{\Gamma_{ln}-i\Omega_\mu}{k_\mu}+\frac{\Gamma_{ml}-i\varepsilon}{k-k_\mu}\right)\zeta\right.\right.$$
$$\left.\left.-\frac{D}{3}\left(\frac{1}{k_\mu}+\frac{1}{k-k_\mu}\right)\zeta^3\right]\right\}^2\Theta(k-k_\mu).$$

The expression for $P_\mu^{(2)}$ in the case $k_\mu < k$ is found from the expression given here by interchanging variables, $\zeta' \leftrightarrow \zeta''$, in the square brackets. When the waves are propagating in opposite directions ($\mathbf{k}_\mu \uparrow\downarrow \mathbf{k}$) the nonlinear interference effect and the field splitting are not manifested ($P_\mu^{(2)} = P_\mu^{(3)} = 0$), and in the expression for the population effect $P_\mu^{(1)}$ we should change the sign of the frequency deviation of the strong field: $\Omega \to -\Omega$.

The factor $1 - A_{mm}/(\Gamma_{mm} + D\zeta^2)$ describes the contribution of spontaneous m–n transitions. If level m decays exclusively to level n ($A_{mn} = \Gamma_{mn}$), then we have $P_\mu^{(1)} = 0$ in the absence of diffusion ($D = 0$); i.e., in level n the external field does not produce a nonequilibrium Bennett structure. This well-known fact is explained on the basis that all the atoms which are put in state m as a result of absorption will necessarily return to level n with the same velocity $\mathbf{v}$. If, however, the atoms undergo a diffusion in velocity space over the lifetime Γ_{mm}^{-1} in level m, this compensation will be disrupted, and a wide nonequilibrium structure of a diffusion nature will arise in level m.

A further simplification occurs in the case $\mathbf{k}_\mu \uparrow\uparrow \mathbf{k}$, $k_\mu \approx k, A_{mn} = 0$, in which expressions (3.5) reduce to single integrals, and the term responsible for the field splitting vanishes:

$$P_\mu^{(1)} \propto \operatorname{Re} I_1 = \operatorname{Re} \int_0^\infty \frac{\exp[-(\Gamma_{ln}+\Gamma_{mn}-i\varepsilon)t - {}^2/_3 Dk^2t^3]}{\Gamma_{nn}+Dk^2t^2}\,dt,$$

$$P_\mu^{(2)} \propto \operatorname{Re} I_2 = \operatorname{Re} \int_0^\infty \frac{\exp[-(\Gamma_{ln}+\Gamma_{mn}-i\varepsilon)t - {}^2/_3 Dk^2t^3]}{\Gamma_{ml}-i\varepsilon+Dk^2t^2}\,dt. \tag{3.6}$$

The two lines are centered at the same frequency, $\Omega_\mu = \Omega$, and are symmetric with respect to $\varepsilon = 0$; their shapes, however, are different. The dependence $P_\mu^{(1)}(\varepsilon)$ is a monotonically decreasing dependence at $\varepsilon > 0$, while the function $P_\mu^{(2)}(\varepsilon)$ has two minima in addition to the maximum at $\varepsilon = 0$.

The factor $(\Gamma_{nn} + Dk^2t^2)^{-1}$ at $\varepsilon = 0$ obviously describes the diffusion during the time spent by the ion in level n, while the term $(2/3)Dk^2t^3$ gives the change in the phase caused by the same factor. It can be seen from the expression for $P_\mu^{(2)}$ that only the changes in the phase and the frequency on the λ–n, m–n, and m–l transitions are important in the term representing the nonlinear interference effect, and there is no manifestation of the change in the population distribution due to diffusion. In the case $\mathbf{k}_\mu = \mathbf{k}$, the cancellation of the Doppler shifts of the absorption and scattering of the photons completely eliminates the role played by the dips in the populations. From expressions (3.5) we see that in the case $k_\mu \neq k$ the population dip of width Γ_{mn} enters with a weight of $(k_\mu - k)$, as it does in the absence of diffusion (Ref. 9, for example). For this purpose we need to change the integration variable: $\zeta'' \to \zeta = \zeta'' - \zeta'$. The part of the argument of the exponential function in $P_\mu^{(2)}$ which is linear in ζ and ζ' takes the form

$$-\left(\frac{\Gamma_{ln}-i\Omega_\mu}{k_\mu}+\frac{\Gamma_{mn}+i\Omega}{k}\right)\zeta' - \left(\frac{\Gamma_{mn}+i\Omega}{k}+\frac{\Gamma_{ml}-i\varepsilon}{k_\mu-k}\right)\zeta.$$

In the absence of collisions ($D = 0$), expressions (3.6) reduce to combinations of Lorentzian functions:

$$I_1 = \frac{1}{\Gamma_{nn}(\Gamma_{ln}+\Gamma_{mn}-i\varepsilon)}, \quad I_2 = \frac{1}{(\Gamma_{ml}-i\varepsilon)(\Gamma_{ln}+\Gamma_{mn}-i\varepsilon)}. \tag{3.7}$$

If the condition

$$\Gamma_{nn}+\Gamma_{ml}=\Gamma_{ln}+\Gamma_{mn} \tag{3.8}$$

also holds, e.g., if the relaxation is purely radiative, then the resultant line $I = I_1 + I_2$ reduces to a single Lorentzian line of width Γ_{ml}. There is an interference extinction of the resonance with a width $\Gamma_{mn} + \Gamma_{ln} \gg \Gamma_{ml}$, and we are left with a resonance with the width of the forbidden transition, Γ_{ml}. We would like to know whether a corresponding extinction occurs when there is a diffusion in velocity space, under the assumption that conditions (3.8) hold.

For a qualitative study of the line $I(\varepsilon)$ we go back to expressions (3.6). An integration over ε shows that the area under the line $\operatorname{Re} I_1(\varepsilon)$ is $\pi\Gamma_{nn}^{-1}$, while that under $\operatorname{Re} I_2(\varepsilon)$ is zero. The integral I_1 behaves in the manner of a Bennett dip in a two-level system.[1,4] The linewidth is given in order of magnitude by

$$\delta \sim \max[\Gamma_a, (Dk^2/\Gamma_a)^{1/2}, (Dk^2)^{1/3}], \quad \Gamma_a = \Gamma_{nn} \approx \Gamma_{ln} + \Gamma_{mn}.$$

The relaxation terms in the argument of the exponential function thus reach values on the order of unity at $t \sim \Gamma_a^{-1}$; the same is true of the diffusion term at $t \sim (Dk^2)^{-1/3}$; and the same is true of the diffusion term in the denominator at $t \sim (\Gamma_a/Dk^2)^{1/2}$. Knowing the shortest of these time scales, we can estimate the linewidth, noting that the integral I_1 is the Fourier transform of the integrand at $\varepsilon = 0$.

In the remote wing of the line ($\delta \ll \varepsilon \ll k\bar{v}$) the integrals I_1 and I_2 are dominated by the region $t \lesssim 1/\varepsilon \ll 1/\delta$, so the diffusion is inconsequential there, and an asymptotic expression can be found from (3.7): $\operatorname{Re} I \sim \Gamma_f/\varepsilon^2$, where the width of the forbidden transition is $\Gamma_f = \Gamma_{ml} \ll \Gamma_a$.

Near the center of the line ($\varepsilon \lesssim \Gamma_f$) the leading term in I is I_2. The denominator of the integrand changes more substantially than the exponential function at smaller values of t. If, in addition, the diffusion is sufficiently intense, i.e., if $(Dk^2/\Gamma_f)^{1/2} \gg \Gamma_a$, then we can set $t = 0$ in the argument of the exponential function, and we find $\operatorname{Re} I \sim \operatorname{Re}[Dk^2(\Gamma_f - i\varepsilon)]^{-1/2}$.

In accordance with the qualitative arguments presented above, the profile of the nonlinear resonance found by numerical calculations (Fig. 2a) has a three-scale behavior. This conclusion can also be drawn analytically, by constructing an asymptotic expansion of the function $I(\varepsilon)$ as $p \to \infty$. For this purpose we rewrite (3.6) as double Laplace integrals [we can also find (3.5) from them, by setting $A_{mn} = 0$ and $k_\mu \approx k$]:

$$I_1 = \int_0^\infty dt \int_0^\infty dt' \exp[-(1+\gamma-ix)t - t' - pS],$$

$$I_2 = \int_0^\infty dt \int_0^\infty dt' \exp[-(1+\gamma-ix)t - (\gamma-ix)t' - pS], \tag{3.9}$$

$$S = {}^2/_3 t^3 + t^2 t', \quad x = \varepsilon/\Gamma_a, \quad \gamma = \Gamma_f/\Gamma_a, \quad p = Dk^2/\Gamma_a^3.$$

The standard version of the multidimensional method of steepest descent cannot be applied directly, however, since

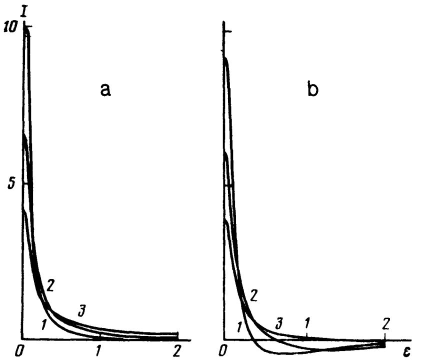

FIG. 2. Profiles of a nonlinear resonance in the interference direction ($\mathbf{k}_\mu \uparrow\uparrow \mathbf{k}$, $k_\mu \approx k$) for $\gamma = 0.1$ and (line 1) $p = 0$, (line 2) 0.1, or (line 3) 1—$A_{mn} = 0$; b—$A_{mn} = \Gamma_{mn}$.

the critical point $t = t' = 0$ of the phase $S(t,t')$ is degenerate. The nature of this degeneracy is illustrated by Fig. 3, from which we see that as we go from positive to negative values of t' the point $t = 0$ converts from a local minimum into a maximum.

The asymptotic form of integrals (3.9) is derived in the Appendix. The results are

$$\Gamma_a^2 \operatorname{Re} I_1 \approx \frac{\pi^{1/2}\Gamma(1/2)}{2p^{1/2}} - \frac{\pi^{1/2}(2/3)^{1/3}\Gamma(2/3)}{p^{2/3}} + O\left(\frac{\ln p}{p}\right), \quad p \gg 1,$$
$$\Gamma_a^2 \operatorname{Re} I_2 \approx \frac{1}{2}\frac{\pi}{(\gamma p)^{1/2}}\left[\frac{1+(1+x^2/\gamma^2)^{1/2}}{2(1+x^2/\gamma^2)}\right]^{1/2} + O(p^{-3/2}), \quad p \gg \gamma. \qquad (3.10)$$

The expression for Re I_2 describes the change in the shape of a narrow peak at the center of the spectrum. Diffusion acts on only the height of this peak, while the half-width at half-maximum, $x_{1/2} = \gamma(3 + 2\sqrt{3})^{1/2} \approx 2.542\gamma$, depends only on the width Γ_{ml} of the forbidden transition. The broad diffusion part has a width $\sim p^{1/2}$ and a height $\sim p^{-1/2}$, so the ratio of the areas under the narrow and wide parts is $\sim (\gamma/p)^{1/2} \ll 1$. At large values of $x \gg p^{1/2}$, the line has a Lorentzian asymptotic behavior $I \propto \gamma/x^2$, so only at the wings of the spectrum do we find an interference extinction of the component of width Γ_{nn}. If spontaneous transitions by the m–n mechanism occur ($A_{mn} \neq 0$), this cancellation at the wings of the line will be disrupted. The function Re $I(\varepsilon)$ then takes on negative values (Fig. 2b).

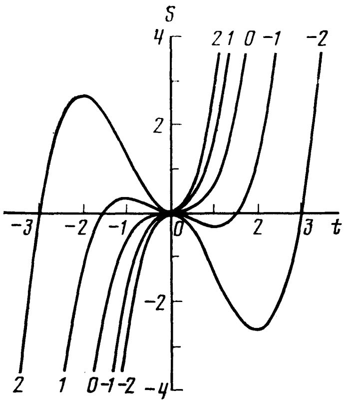

FIG. 3. The phase S of integrals (3.9) versus the variable t at fixed values of t' (the curve labels).

The field on the l–n transition (Fig. 1) has been assumed to be weak. Experimentally, however, it is sometimes convenient to observe nonlinear interference effects in the frequency dependence of the test-field generation power. To calculate this dependence, we should retain in the expression for the work performed by the field, (3.2), not only (3.3) but also terms proportional to the square of the intensity of the test field, $|G_\mu|^4$. Equating the amplification to the loss, we find the shape of the resonance in the case in which both fields are represented by standing waves:

$$|G_\mu|^2 = \frac{\alpha - \beta|G|^2 - \rho}{r}, \quad \alpha = \sum_{\sigma=\pm 1} \langle f_{ln}^\sigma N_{ln} \rangle,$$
$$\beta = \sum_{\sigma=\pm 1} \langle f_{ln}^\sigma f_{nn}(1 - f_{mm}A_{mn})(f_{mn}^{-\sigma} + \bar{f}_{mn}^{\sigma}) N_{mn} \rangle$$
$$+ \sum_{\sigma=\pm 1} \langle f_{ln}^\sigma f_{ml}^\sigma (f_{mn}^\sigma N_{mn} + f_{ln}^\sigma N_{ln}) \rangle, \qquad (3.11)$$
$$r = \sum_{\sigma=\pm 1} \langle f_{ln}^\sigma [f_{ll} + f_{nn}(1 - f_{ll}A_{ln})](f_{ln}^\sigma + \bar{f}_{ln}^{-\sigma}) \rangle,$$

where ρ is the loss, the angle brackets mean a convolution over all of the intermediate arguments with a δ-function [as in (3.3)], and the Green's functions which correspond to waves traveling in the positive and negative directions are found from general expression (2.12):

$$f_{ij}^{\pm}(\xi|\xi'; \Gamma, \mathbf{p}) = f_{ij}(\xi|\xi'; \Gamma, \pm\mathbf{p}). \qquad (3.12)$$

In the absence of a strong field, expression (3.11) determines the shape of the ordinary Lamb dip. The term β introduces an additional dependence on Ω_μ: the resonances at $\Omega_\mu = \pm\Omega$ which were discussed above.

4. POLARIZATION EFFECTS

In the model of nondegenerate states, the narrow nonlinear-interference peak in the work performed by the test field and the broad populated resonance coalesce. To obtain more-detailed information about the medium, we would like to separate resonances which differ in nature. In the case of a two-level system this can be done by the methods in polarization spectroscopy,[9,11] by choosing the polarizations of the strong and test waves in such a way that one effect or another in the spectrum is emphasized. We would like to see whether it is possible to separate the narrow and broad resonances by means of polarization effects.

We start with an expression for the work performed by the field of the test wave in this case in a scheme of Raman scattering through the upper level (Fig. 1b), ignoring radiative decay by the m–l and m–n mechanisms[9]:

$$P_\mu = -2\hbar\omega_\mu \operatorname{Re}\Big\{ N_{ml}\alpha_{ml}|G_\mu|^2 - N_{mn}\sum_{\varkappa q}\big[|J(\varkappa q)|^2\beta_\varkappa$$
$$+ I_\mu(\varkappa q) I^*(\varkappa q) B_\varkappa\big]\Big\}, \qquad (4.1)$$
$$G_{\mu\sigma} = -E_{\mu\sigma} d_{ml}/2\hbar, \quad |G_\mu|^2 = \sum_\sigma |G_{\mu\sigma}|^2,$$
$$\alpha_{ml} = \langle f_{ml1} \rangle, \quad N_{ij} = \frac{r_{ii}}{2J_i+1} - \frac{r_{jj}}{2J_j+1},$$
$$\beta_\varkappa = a_{\beta\varkappa} A_{\beta\varkappa}, \quad B_\varkappa = a_{B\varkappa} A_{B\varkappa};$$
$$A_{\beta\varkappa} = \langle f_{ml1}(0|\xi_1) f_{mn\varkappa}(\xi_1|\xi_2)(f_{mm}(\xi_2|0) + \bar{f}_{mn1}(\xi_2|0)) \rangle,$$
$$A_{B\varkappa} = \langle f_{ml1}(0|\xi_1) f_{nl\varkappa}(\xi_1|\xi_2)(f_{ml1}(\xi_2|0) + f_{mn1}(\xi_2|0)) \rangle.$$

TABLE I. Polarization coefficients α and a for various combinations of the angular momenta of the levels for four polarization states of the strong and test waves.

J_n	J_m	J_l	$\alpha_{\uparrow\uparrow}=a_{\uparrow\uparrow}$	$\alpha_{\uparrow\rightarrow}=a_{\uparrow\rightarrow}$	$\alpha_{++}=a_{++}$	$\alpha_{+-}=a_{+-}$
J	$J-1$	$J-2$	$\frac{6}{5}\frac{1}{2J-1}$	$\frac{9}{10}\frac{1}{2J-1}$	$\frac{3}{10}\frac{1}{2J-1}$	$\frac{3}{10}\frac{1}{2J-1}$
J	$J-1$	$J-1$	$\frac{3}{5}\frac{J+1}{J(2J-1)}$	$\frac{3}{10}\frac{4J-1}{J(2J-1)}$	$\frac{3}{10}\frac{3J-2}{J(2J-1)}$	$-\frac{3}{10}\frac{2J-3}{J(2J-1)}$
J	$J-1$	J	$\frac{3}{5}\frac{4J^2+1}{J(4J^2-1)}$	$\frac{3}{10}\frac{(6J-1)(J+1)}{J(4J^2-1)}$	$\frac{3}{5}\frac{6J^2-1}{J(4J^2-1)}$	$\frac{3}{10}\frac{(J-1)(2J-3)}{J(4J^2-1)}$
J	J	$J-1$	$\frac{3}{5}\frac{J-1}{J(2J+1)}$	$\frac{3}{10}\frac{4J+1}{J(2J+1)}$	$\frac{3}{10}\frac{2J+3}{J(2J+1)}$	$-\frac{3}{10}\frac{2J+3}{J(2J+1)}$
J	J	J	$\frac{3}{5}\frac{3J^2+3J+1}{J(J+1)(2J+1)}$	$\frac{3}{10}\frac{2J^2+2J+1}{J(J+1)(2J+1)}$	$\frac{3}{5}\frac{2J^2+2J+1}{J(J+1)(2J+1)}$	$\frac{3}{5}\frac{(2J-1)(2J+3)}{J(J+1)(2J+1)}$
J	$J+1$	J	$\frac{3}{5}\frac{4J^2+8J+5}{(J+1)(2J+1)(2J+3)}$	$\frac{3}{10}\frac{J(6J+7)}{(J+1)(2J+1)(2J+3)}$	$\frac{3}{5}\frac{6J^2+12J+10}{(J+1)(2J+1)(2J+3)}$	$\frac{3}{10}\frac{(J+2)(2J+3)}{(J+1)(2J+1)(2J+3)}$

$$a_{\beta\varkappa}=9\begin{Bmatrix}1 & \varkappa & 1\\ J_l & J_m & J_n\end{Bmatrix}^2,$$

$$a_{B\varkappa}=(-1)^{J_n-J_l}\begin{Bmatrix}1 & \varkappa & 1\\ J_m & J_l & J_m\end{Bmatrix}\begin{Bmatrix}1 & \varkappa & 1\\ J_m & J_n & J_m\end{Bmatrix}; \quad (4.2)$$

$$J(\varkappa q)=\sum_{\sigma\sigma_1}(-1)^{1-\sigma_1}\langle 1\sigma 1-\sigma_1|\varkappa q\rangle G_{\mu\sigma}G_{\sigma_1}^*,$$

$$I_\mu(\varkappa q)=\sum_{\sigma\sigma_1}(-1)^{1-\sigma_1}\langle 1\sigma 1-\sigma_1|\varkappa q\rangle G_{\mu\sigma}G_{\mu\sigma_1}^*, \quad (4.3)$$

$$I(\varkappa q)=\sum_{\sigma\sigma_1}(-1)^{1-\sigma_1}\langle 1\sigma 1-\sigma_1|\varkappa q\rangle G_\sigma G_{\sigma_1}^*.$$

Here $\beta_\varkappa$ describes the nonlinear interference, and $B_\varkappa$ describes the saturation effect. The functions $A_{\beta\varkappa}(\Omega_\mu)$, and $A_{B\varkappa}(\Omega_\mu)$ specify the spectral dependence and the dependence on $\varkappa$; the polarization dependences are in the polarization tensors J, I_μ, and I and in the coefficient $a_{\beta\varkappa}$ and $a_{B\varkappa}$.

In the simplest case, in which the spectral functions $A_{\beta\varkappa}$ and $A_{B\varkappa}$ do not depend on $\varkappa$, the nonlinear part of the work performed by the field is

$$\Delta P_\mu=2\hbar\omega_\mu N_{nm}\,\mathrm{Re}(\alpha A_\beta(\Omega_\mu)+aA_B(\Omega_\mu)),$$
$$\alpha=\sum_{\varkappa q}I_\mu(\varkappa q)I^*(\varkappa q)a_{\beta\varkappa},\quad a=\sum_{\varkappa q}|J(\varkappa q)|^2a_{B\varkappa}. \quad (4.4)$$

We can write expressions for α and a in the cases of parallel ($\uparrow\uparrow$) and orthogonal ($\uparrow\rightarrow$) linear polarizations of the saturating and test fields and also for the cases of the same ($+\,+$) and the opposite ($+\,-$) circular polarizations of these fields:

$$\alpha_{\uparrow\uparrow}=\frac{a_{\beta0}+2a_{\beta2}}{3},\quad a_{\uparrow\uparrow}=\frac{a_{B0}+2a_{B2}}{3},$$
$$\alpha_{\uparrow\rightarrow}=\frac{a_{\beta1}+a_{\beta2}}{2},\quad a_{\uparrow\rightarrow}=\frac{a_{B0}-a_{B2}}{3},$$
$$\alpha_{++}=\frac{2a_{\beta0}+3a_{\beta1}+a_{\beta2}}{6},\quad a_{++}=\frac{2a_{B0}+3a_{B1}+a_{B2}}{6}, \quad (4.5)$$
$$\alpha_{+-}=\frac{2a_{\beta0}-3a_{\beta1}+a_{\beta2}}{6},\quad a_{+-}=a_{B2}.$$

Let us check whether the values of α and a are the same for each of these simplest polarization states. For this purpose we introduce $J_n=J$, and we write all possible combinations of the angular momenta (J_m, J_l) which are allowed by the selection rules for the dipole moment in a three-level system: $(J-1, J-2)$, $(J-1, J-1)$, $(J-1, J)$, $(J, J-1)$, (J, J), and $(J+1, J)$. There are a total of six such independent combinations; other combinations are found through the interchange $n\leftrightarrow l$, under which expressions (4.2) are symmetric. Direct calculations yield equal values for the coefficients α and a for all possible combinations of angular momenta (Table I).

It is natural to suggest that the coefficients α and a are equal for arbitrary polarizations. To prove this suggestion we rewrite expression (4.2) for $a_{B\varkappa}$, using Eq. (108.9) from Ref. 12:

$$a_{B\varkappa}=(-1)^\varkappa\sum_j(-1)^j(2j+1)a_{\beta j}\begin{Bmatrix}1 & \varkappa & 1\\ 1 & j & 1\end{Bmatrix}. \quad (4.6)$$

Now making use of the relation between the $6j$ and $3j$ symbols [Eq. (108.4)] and the orthogonality condition for the $3j$ symbols [Eq. (106.13)], we can show that the relation $a=\alpha$ holds.

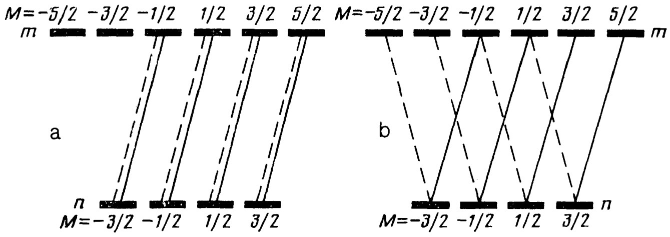

FIG. 4. Transition schemes in a two-level system with $J_m=5/2$ and $J_n=3/2$. The strong field (the solid line) has a right-handed circular polarization, while the test field (dashed lines) has (a) a right-handed or (b) left-handed polarization. The quantization axis for the angular momenta is chosen along the direction of the wave vector $\mathbf{k}_\mu\uparrow\uparrow\mathbf{k}$.

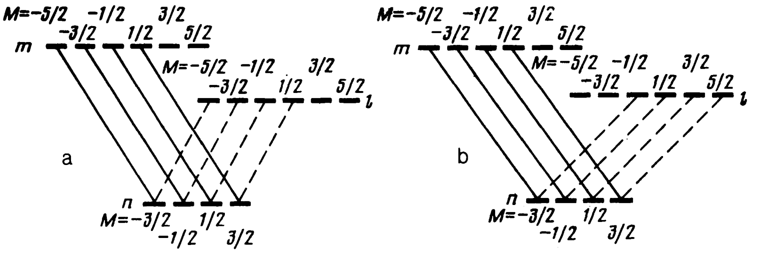

FIG. 5. Scheme of transitions in a three-level system in the case of a Raman scattering through the lower level with $J_n = 3/2$ and $J_m = J_l = 5/2$. The strong field (solid lines) has a left-handed circular polarization, while the test field (dashed lines) has (a) a coincident polarization or (b) an orthogonal polarization.

The relation between the resultant amplitude of the population-associated and nonlinear interference terms is thus independent of the polarization of the radiation and remains the same as in the model of nondegenerate states. The particular state of the polarizations of the strong and test fields influences only the common factor (Table I), in contrast with the case of a two-level system, in which the shape of a nonlinear resonance may change radically in the transition from identically polarized fields to orthogonally polarized fields (Chapter 5 in Ref. 9).

The physical reason for the difference between the polarization effects in two-level and three-level systems can be explained qualitatively in a simple way. When the polarizations of the fields are the same, a two-level system breaks up into a set of two-level subsystems (Fig. 4a), but when the polarizations are orthogonal it breaks up into a set of three-level subsystems (Fig. 4b). In the three-level subsystems, other effects may become substantial; in particular, there may be effects of resonances with the width of the forbidden transition, whose role in this case is played by a transition between magnetic sublevels of the same level. In the case of a three-level system in contrast, there is no qualitative change in the scheme of transitions when we switch from one state of the polarizations to another. The example in Fig. 5 illustrates that when coincident polarizations of the strong and test fields are replaced by orthogonal polarizations the only changes which occur are in the indices of the working sublevels.

5. DISCUSSION

The results derived above, which determine the shape of a nonlinear resonance, lead to the conclusion that studying the broadening of the central peak is not a good way to study diffusion in velocity space. The half-width at half-maximum of such a peak is about 2.5 times the relaxation constant of the forbidden transition, Γ_{ml}, and does not depend on the diffusion coefficient D. Information on diffusion can be extracted either at the height of the central peak, $\propto p^{-1/2}$, or from the broad diffusion part of the lineshape.

As an example we consider a low-temperature plasma in which the Coulomb scattering of excited ions by ground-state ions is associated with a random walk of the excited ions in velocity space:

$$D=\frac{\nu_{ii}\bar{v}_i^2}{2},\quad \nu_{ii}=\frac{16\pi^{1/2}Le^4Z^2}{3m^2\bar{v}_i^3}\sum_{Z'}N_{Z'}Z'^2. \qquad (5.1)$$

Here $N_{Z'}$ is the density of perturbing ions, with a charge $Z'e$; Ze is the charge of the radiating ion; m and $\bar{v}_i$ are the mass and thermal velocity of the ions; and L is the Coulomb logarithm.

Let us estimate the diffusion parameter p for the plasma of an ion laser working in the visible part of the spectrum ($Z = Z' = 1$). For this estimate we will need the relaxation constants of the levels and the plasma parameters. Table II lists the eight strongest lines in the output of an argon laser, which correspond to transitions between the $4p$ and $4s$ configurations of the ArII ions. The radiative-relaxation constants were taken from the review in Ref. 13; here $\Gamma_{ij} = \gamma_i + \gamma$. We see from this table that the upper levels $4p^2D^0_{5/2}$ and $4p^2P^0_{3/2}$ are common, and the following three-level systems with a Λ configuration can be constructed (Fig. 1b): 4545/4765 and 4727/4965 Å. The low-lying states $4s^2P_{3/2}$ and $4s^2P_{1/2}$ are common to several transitions, so the lines listed in Table I can be used to construct 13 two-level systems with a V configuration (Fig. 1a). In particular, the strong lines 4880/5145 Å are coupled through the lower level. This is the pair of lines which was used in the experiments by Lebedeva, Odintsov, *et al.*[8]

TABLE II. Radiative relaxation constants of the levels for the lines in the output of an ArII ion laser.

λ, Å	Upper level m	Lower level n	$\nu_m \times 10^{-7}\,s^{-1}$	$\nu_n \times 10^{-9}\,s^{-1}$
4545	$4p^2P^0_{3/2}$	$4s^2P_{3/2}$	6	1,25
4579	$4p^2S^0_{1/2}$	$4s^2P_{1/2}$	7	1,25
4658	$4p^2P^0_{1/2}$	$4s^2P_{3/2}$	6	1,25
4727	$4p^2D^0_{3/2}$	$4s^2P_{3/2}$	6	1.25
4765	$4p^2P^0_{3/2}$	$4s^2P_{1/2}$	6	1.25
4880	$4p^2D^0_{5/2}$	$4s^2P_{3/2}$	6	1.25
4965	$4p^2D^0_{3/2}$	$4s^2P_{1/2}$	6	1.25
5145	$4p^4D^0_{5/2}$	$4s^2P_{3/2}$	8	1,25

The electron density in the plasma of an argon laser is typically $n_e \sim 10^4$ cm^{-3}, and the average velocity is $\bar{v}_i \sim 10^5$ cm/s. From (5.1) we find $\nu_{ii} \sim 3\times 10^7$ s^{-1} and $D \sim 4\times 10^{17}$ cm^2/s. Using γ_n from Table II, we then find $p\sim 1 \gg \gamma \sim 10^{-1}$. Under these conditions one can thus observe a narrow peak against the background of a wide line. Most of the diffusion effect is in the square-root dependence of the amplitude of the sharp resonance on the collision rate.

In Ref. 8 the measured width of the resonance in the output, extrapolated to zero values of the density n_e and the saturating field G, was 67 ± 5 MHz, in the case in which the test field interacted with the $4p^2D^0_{5/2} - 4s^2P_{3/2}$ transition; it was 130 ± 10 MHz when the $4p^4D^0_{5/2} - 4s^2P_{3/2}$ transition was probed. The calculated width of the forbidden transition is $\Delta\nu_{ml} \approx 40$ MHz; it varies by less than 10% if we use the data in Table II (calculations by Tang *et al.*; cited in the review in Ref. 13) and the measurements by Bennett *et al.*[14] or the calculations by Loginov and Gruzdev.[15] The measured width is two or three times $\Delta\nu_{ml}$; this result can be explained in terms of a distortion of the Lorentzian shape of the resonance by Coulomb diffusion. The observed dependence of the width on the electron density may be related to either Stark broadening or decay of the m and l levels as a result of inelastic collisions of the ion with electrons. Resolving these questions will require measurements of the dependence of the shape of the resonance on the plasma parameters.

The abundance of possible pairs of convenient adjacent transitions, the difference of more than an order of magnitude in the relaxation constants of the upper and lower levels, the combination of measurements in interference and noninterference directions, and the combining of Λ- and V-shaped schemes—all these favorable circumstances qualify the plasma of an argon laser as a convenient system for the development of methods of nonlinear spectroscopy and their application to plasma diagnostics.

We wish to thank S. A. Babin and A. M. Shalagin for useful discussions.

APPENDIX

The mathematical theory of the asymptotic form of the Laplace integral with a degenerate critical point is set forth in detail in Chapter II of the book by Arnol'd *et al.*[16] To calculate the expansion coefficients, we borrow a method from that book: We assume that the phase S is one of the new variables. The integral then transforms into a Laplace transform of the integral over the remaining variable (the integrand in the last integral is called a "Gel'fand-Leray form"). The asymptotic form of the Laplace transform is found from

$$\int_0^\infty e^{-ps} s^\alpha (\ln s)^k \, ds \approx \frac{d^k}{d\alpha^k} \frac{\Gamma(\alpha+1)}{p^{\alpha+1}}, \quad p\to\infty. \tag{A1}$$

A difficulty arises in the switch from the variables (t,t') to the variables (s,ξ): A single-valued transformation cannot be made because of the degenerate nature of the critical point. It is necessary to construct a resolution of the singularity. For this purpose we use the σ-process (see Sec. 2 in Ref. 17), which reduces in the case at hand to a preliminary replacement of (t,t) by (t,v) where $v = t'/t + 2/3$.

In terms of the new variables we have

$$I_i = \int_0^\infty ds\, e^{-ps} J_i(s),$$

and the integrals J_i of the Gel'fand-Leray forms reduce to

$$J_1(s) = \frac{1}{2} s^{-1/3} \int_{\xi_0}^\infty d\xi\, \xi^{-1/3} \exp\left[-s^{1/3}\left(\xi + \frac{1/3+\gamma-ix}{\xi^{1/2}} \right)\right],$$

$$J_2(s) = \frac{1}{2} s^{-1/3} \int_{\xi_0}^\infty d\xi\, \xi^{-1/3} \cdot \exp\left[-s^{1/3}\left(\xi(\gamma-ix) + \frac{1+\gamma/3-ix/3}{\xi^{1/2}} \right)\right], \tag{A2}$$

where $\xi = v^{2/3}$ and $\xi_0 = (2/3)^{2/3}$. Expanding (A2) in a series of incomplete Γ functions,

$$J_1 = \sum_{k=0}^\infty \frac{(-1)^k}{k!} \Gamma\left(\frac{1-k}{2}, \xi_0 s^{1/3} \right) s^{k/2} \left(\frac{1}{3} + \gamma - ix \right)^k,$$

$$J_2 = \sum_{k=0}^\infty \frac{(-1)^k}{k!} \Gamma\left(\frac{1-k}{2}, \xi_0 s^{1/3} (\gamma-ix) \right) \cdot s^{k/2} (\gamma-ix)^{(k-1)/2} \left(1 + \frac{\gamma-ix}{3} \right)^k,$$

$$\Gamma(a,x) = \int_a^\infty \xi^{a-1} e^{-\xi}\, d\xi,$$

and using power series for the Γ functions[18] and relation (A1), we find (3.10): the leading terms of the real part of the asymptotic behavior. The degenerate nature of the critical point of the phase is seen in the circumstance that in addition to the powers $p^{-n/2}$ $(n = 1,2,\ldots)$ the expansion contains terms $\propto p^{-k/3}$ $(k = 2,3,\ldots)$ and also $\ln p$. For this reason, the terms of the series decrease extremely slowly. Comparing the first term of the expansion with the second, we see that the condition under which we can approximate the expansion by the leading term is $p \gg 1$ for I_1 and $p \gg \gamma$ for I_2. In the case $\gamma \ll 1$, it is sufficient to restrict the expansion for I_2 to the first term, even at $p\sim 1$ (line 3 in Fig. 2).

1) The collisional shifts Δ_{ij} of the resonant transition frequency can be dealt with in Eqs. (2.5) and below through the replacement $\Gamma_{ij} \to \Gamma_{ij} + i\Delta_{ij}$.
2) The expression for $P_\mu^{(3)}$ is the same as that derived by Berman,[10] who examined that expression alone.

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