Gas-susceptibility saturation in a standing-wave field following multiphoton excitation near resonance with intermediate levels

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The problem of q-photon interaction between a $(q+1)$-level atom and electromagnetic waves that are at resonance with neighboring atomic transitions is solved. It is shown that if the initial and final levels are metastable while the intermediate states decay rapidly and only radiatively, a transition to a generalized two-level system (GTS) of metastable levels is possible. A new derivation is presented of the GTS density matrix and of expressions for the system response. The influence of the radiative transitions between levels is considered and it is shown that these transitions must in principle be taken into account near resonance with intermediate levels. The relations derived are used to calculate the linear interaction between a gas of multilevel atoms and a standing-wave field. In this interaction the line broadening due to the linear Doppler effect is completely eliminated and a multiphoton resonance sets in, having a width on the order of the width of the transition between the metastable levels. The effect exerted on this resonance by spatial modulation of the medium in the field of an intense standing wave is analyzed. It is shown that with increasing saturation parameter the line width first increases linearly and the shape ceases to depend on the relaxation processes on the metastable levels. However, the singularities in the gas-susceptibility derivatives, which lead to the onset of lines free of field-induced broadening, are preserved. The feasibility of observing the narrow resonance and using it in atomic spectroscopy is discussed.

1. INTRODUCTION

The basic results of high-resolution spectroscopy were obtained using one-photon resonances and two-photon absorption resonances. Yet the availability of tunable lasers permits also an effective use of multiphoton processes, since the photon frequencies can then be tuned to resonance with an intermediate level. These processes permit the investigation of forbidden transitions as well as of transitions in the short-wave and other bands, for which there is no resonant radiation. The ensuing possibilities are considered in the present paper.

It is known, with the three-level system as the example,[1] that the width of a multiphoton line is of the order of the width γ of the transition between the initial level 0 and the final one q. The Doppler broadening ω_D is eliminated only in part near a resonance with an intermediate level, and is of the order of[2–4] the rate Γ of decay of these levels. In the situation of greatest interest for spectroscopy, when the levels 0 and q are metestable,

$$\gamma/\Gamma \ll 1, \tag{1}$$

the broadening is still large. In Refs. 5 and 6 are advanced theoretical arguments that this broadening of resonance, due to the nonlinear reaction with a standing-wave field, does not occur in a gas of three-level atoms.[1)] It will be shown here that such an effect occurs also for a multiphoton line. It follows that nonlinear interaction with a standing wave is a universal method of eliminating Dopler broadening, equally suitable for an ordinary two-level medium, in which the Lamb dip is observed, and in an arbitrary mulitlevel case.

The condition (1) is known from the Lamb-dip theory. It is used to confirm the validity of the rate approximation[2)] (Ref. 9) and to take approximately into account the spatial modulation of the medium.[11] In the two-level case, however, Eq. (1) is satisfied only for strongly differing level-decay rates or for large collision broadening of the line. For the lines used in one-photon precision spectroscopy we have $\gamma \sim \Gamma$, and here the accuracy of the rate approximation is 30% (see the results of the numerical calculations in Ref. 12). In our case (1) is always satisfied. The rate approximation is thus not applicable at all, and the effect of spatial modulation plays the principal role even in a weak field,[13] and leads in the case of strong saturation to the line deformation considered in the present paper.

A formalism convenient for the description of multiphoton processes is that of the generalized two-level system (GTS). It was developed in Refs. 14 and 15 for the nonresonant case. The presence of the small parameter (1) makes the use of the method possible in the case of resonances with intermediate levels. In this case the effective Hamiltonian and the generalized dipole-moment matrix increase resonantly, and the expressions obtained for them in Refs. 14 and 15 no longer hold. We present below a calculation of these quantities by a new procedure.

2. EFFECTIVE HAMILTONIAN

We consider a $(q+1)$-level atom in the field of q resonant waves

$$\mathscr{E} = \sum_{j=-q}^{q} \mathscr{E}_j \exp(-i\omega_j t), \tag{2}$$

where $j \neq 0$. At $j > 0$, $\mathscr{E}_j$ and ω_j are the amplitude and the frequency of the wave that is at resonance with the transition

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between the levels $j-1$ and j, $\mathscr{E}_{-j}=\mathscr{E}_j^*$, $\omega_{-j}=-\omega_j$. We are interested in a situation in which the initial and final levels 0 and q are metastable, while the intermediate levels decay rapidly. Since the collision broadening of the levels is of the same order, such a situation is realized in the pressure range in which the intermediate levels are only radiatively broadened. We have then for the evolution of the density matrix $\bar{\sigma}$ during a time shorter than γ^{-1} the equation

$$i(\dot{\bar{\sigma}}+\{\Gamma,\bar{\sigma}\})=[\bar{V},\bar{\sigma}], \tag{3}$$

where $\{\Gamma,\bar{\sigma}\}=\Gamma\bar{\sigma}+\bar{\sigma}\Gamma$, $\Gamma_{ij}=\delta_{ij}\Gamma_i$, $2\Gamma_i$ is the rate of decay of the ith level, $\Gamma_0=\Gamma_q=0$, $\bar{V}=-d\mathscr{E}$ is the Hamiltonian of the interaction of the atom with the field (2), and d is the dipole-moment operator. We consider below a cascade level scheme $\omega_{j,j-1}>0$ with ω_{ij} the frequency of the $i\to j$ transition. We obtain for it

$$\bar{V}_{j,\,j-1}=G_j\exp(-i\Omega_jt),\qquad \bar{V}_{j-1,\,j}=G_{-j}\exp(i\Omega_jt), \tag{4}$$

where $G_j=G^*_{-j}=-d_{j,j-1}\mathscr{E}_j$, $d_{m,n}$ is the dipole matrix element of the transition $m\to n$, $\Omega_j=\omega_j-\omega_{j,j-1}$ is the detuning of the frequency of the j-th field. In the case of a bent level scheme it is necessary to make in (4) the substitutions $\mathscr{E}_j\to\mathscr{E}_{-j}$, $\omega_j\to\omega_{-j}$ for transitions in which $\omega_{j,j-1}<0$. We seek the solution of (3) in the form

$$\bar{\sigma}=\exp(-\Gamma t)\sigma\exp(-\Gamma t).$$

We then obtain

$$i\dot{\sigma}=V(t)\sigma-\sigma V^+(t), \tag{5}$$

where $V(t)=\exp(\Gamma t)\bar{V}\exp(-\Gamma t)$.

Let the condition for q-photon resonance be satisfied in the system, viz., the total detuning of the frequency of $\nu=\Omega_1+\cdots+\Omega_q$ is small compared with Γ. Then, considering the evolution of σ after a time

$$\Gamma^{-1}\ll T\ll\nu^{-1}, \tag{6}$$

we get

$$\sigma(t+T)=S(T)\sigma(t)S^+(T),$$
$$S(T)=\sum_{n=0}^{\infty}S_n,\qquad S_n=(-i)^n\int_t^{t+T}dt_1\ldots\int_t^{t_{n-1}}dt_nV(t_1)\ldots V(t_n). \tag{7}$$

We introduce the metastable-levels density matrix $\rho_{ik}=\sigma_{i,k}$, where the subscripts i and k are equal to 0 or 1. We are interested in the connection between $\rho(t)$ and $\rho(t+T)$. Assume that at the instant t we have

$$\sigma_{ik}=\begin{cases}\rho_{ik} & \text{at } i,k, \text{ equal to 0 or } q\\ 0 & \text{in the remaining cases}\,.\end{cases} \tag{8}$$

When (6) is satisfied, the main term in $\rho(t+T)$ is the one linear in T. The terms independent of T contain the parameter $(\Gamma T)^{-1}$, and the terms of higher powers in T contain the parameter νT. The term linear in T appears in the q-th term of the expansion of $\rho(t+T)$ in terms of the field

$$\rho_q=\sum_{n=0}^{q}S_n\rho(t)S^+_{q-n}. \tag{9}$$

In (9), $S_n\sim(G/\Gamma)^n$ at $n\neq q$ and $S_q=(G/\Gamma)^q\Gamma T$. We see thus that only the terms with $n=0$ or q need be retained in (9). We then obtain

$$i\dot{\rho}=U\rho-\rho U^+, \tag{10}$$

where

$$\dot{\rho}=\rho_q/T,\qquad U=iS_q/T \tag{11}$$

is the effective GTS Hamiltonian. Terms linear in T occur in all the terms of order $q+2n$ of the field expansion of $\rho(t+T)$, with n an integer. If

$$G_j/\Gamma\ll1, \tag{12}$$

they are small compared with those taken into account in (9). All the quantities will hereafter be calculated in the lowest order in G/Γ. For the matrix elements of U we obtain from (7)

$$U_{0q}=\exp(i\nu t)G_1^*\ldots G_q^*[(\Sigma_1+i\Gamma_1)\ldots(\Sigma_{q-1}+i\Gamma_{q-1})]^{-1},$$
$$U_{q0}=\exp(-i\nu t)G_1\ldots G_q[(\Sigma_1+i\Gamma_1)\ldots(\Sigma_{q-1}+i\Gamma_{q-1})]^{-1}, \tag{13}$$
$$U_{00}=|G_1|^2/(\Sigma_1+i\Gamma_1),\qquad U_{qq}=|G_q|^2/(\Sigma_{q-1}+i\Gamma_{q-1}),$$

where $\Sigma_s=\Omega_1+\ldots+\Omega_s$.

3. GENERALIZED DIPOLE MOMENT MATRIX

Let the atom be now situated in a field

$$\mathscr{E}=\sum_j\lambda_j\mathscr{E}_j\exp(-i\omega_jt).$$

From the expression $P(t)=\mathrm{Sp}(d\bar{\sigma}(t))$ for the polarization of the atom we get for the response at the frequency ω_j

$$P_j=\int_t^{t+T}\frac{d\tau}{T}\exp(i\omega_j\tau)P(\tau)$$

and taking the boundary condition (8)

$$P_j=\mathrm{Sp}(D_j\rho), \tag{14}$$

where

$$D_j=-\mathscr{E}_{-j}^{-1}A_{-j} \tag{15}$$

is the generalized dipole-moment matrix, and

$$A_j=\int_t^{t+T}\frac{d\tau}{T}S^+(\tau-t)\exp(-2\Gamma\tau)\frac{\partial V(\tau)}{\partial\lambda_j}S(\tau-t). \tag{16}$$

Here and below, the derivatives are calculated at $\lambda_j=1$. That D is Hermitian $(D_{-j}=D_j^+)$ follows from the identity

$$\exp(-2\Gamma\tau)V(\tau)=V^+(\tau)\exp(-2\Gamma\tau). \tag{17}$$

From the equations for $\partial S/\partial\lambda_j$ we obtain

$$\frac{\partial V(\tau)}{\partial\lambda_j}S(\tau-t)=iS(\tau-t)\frac{d}{d\tau}\left[S^{-1}(\tau-t)\frac{\partial S(\tau-t)}{\partial\lambda_j}\right].$$

Substituting this expression in (16) and integrating by parts, we obtain, taking (17) into account,

$$A_j=\frac{i}{T}S^+(T)e^{-2\Gamma(t+T)}\frac{\partial S(T)}{\partial\lambda_j}$$

$$+2i\int_t^{t+T}\frac{d\tau}{T}S^+(\tau-t)\Gamma e^{-2\Gamma\tau}\frac{\partial S(\tau-t)}{\partial\lambda_j}. \qquad (18)$$

That part of A_j which is independent of T arises in the g-th order in the field. Expanding in analogy with (9) the first term of (18) and recognizing that $\partial S_0/\partial\lambda_j=0$, we get ultimately

$$A_j=\frac{\partial U}{\partial\lambda_j}+2i\int_t^{t+T}\frac{d\tau}{T}S^+(\tau-t)\Gamma\exp(-2\Gamma\tau)\frac{\partial S(\tau-t)}{\partial\lambda_j}. \qquad (19)$$

The matrix elements D at $\Gamma=0$ were calculated in Ref. 15, and sums over the intermediate levels appear in them if $\Gamma\neq 0$. If each photon is resonant, their calculation is elementary.[16] We then obtain

$$(A_j)_{q,0}=\theta(j)\exp(-i\nu t)G_1\ldots G_q\cdot$$
$$\cdot[(\Sigma_1+i\Gamma_1)\ldots(\Sigma_{j-1}+i\Gamma_{j-1})(\Sigma_j-i\Gamma_j)\ldots(\Sigma_{q-1}-i\Gamma_{q-1})]^{-1},$$
$$(A_j)_{0,0}=(1-\delta_{|j|,q})|G_1\ldots G_j|^2[(\Sigma_1^2+\Gamma_1^2)\ldots(\Sigma_{|j|-1}^2+\Gamma_{|j|-1}^2)$$
$$\times(\Sigma_{|j|}-i\,\mathrm{sign}(j)\Gamma_{|j|})]^{-1}, \qquad (20)$$
$$(A_j)_{q,q}=(1-\delta_{|j|,1})|G_j\ldots G_q|^2[(\Sigma_{q-1}^2+\Gamma_{q-1}^2)\ldots(\Sigma_{|j|}^2+\Gamma_{|j|}^2)$$
$$(\Sigma_{|j|-1}+i\,\mathrm{sign}(j)\Gamma_{|j|-1})],$$

where

$$\theta(x)=\begin{cases}1 & \text{at } x>0,\\ 0 & \text{at } x<0.\end{cases}$$

4. ALLOWANCE FOR RADIATIVE TRANSITIONS BETWEEN LEVELS

If the products of the radiative decay include levels 0,..., q, we obtain in lieu of (3) for the diagonal elements

$$\dot{\bar\sigma}_{jj}+2\Gamma_j\bar\sigma_{jj}-\sum_l\Gamma_{lj}\bar\sigma_{ll}=-i[\bar V,\bar\sigma]_{jj}, \qquad (21)$$

where Γ_{lj} is the rate of the radiative transition $l\to j$.

Equations (21) are not Hamiltonian and the above solution method cannot be used. In multiphoton interaction, however, the real populations of the intermediate levels are small. In the zeroth approximation in $\bar\sigma_{jj}$ we can disregard completely the equations for them, or leave them the same as in (3). Then, substituting the solution of (3) in the right-hand side of (21) we can calculate $\bar\sigma_{jj}$ and the terms, proportional to them, of the arrival at the metastable levels. The results are the following final equations for the GTS density matrix[16]:

$$\frac{\partial\rho_{i,k}}{\partial t}=-i(U\rho-\rho U^+)_{i,k}+2\delta_{ik}\,\mathrm{Im}\left\{\sum_{j=1}^{q-1}w_{j,i}\right.$$
$$\left.\cdot[(A_j-A_{j+1})_{q0}\rho_{0q}+\delta_{1,j}(A_1)_{00}\rho_{00}-\delta_{q-1,j}(A_q)_{qq}\rho_{qq}]\right\}, \qquad (22)$$

where

$$w_{j,i}=\sum_{(j_1,\ldots j_m)}w_{j,i}(j_1\ldots j_m) \qquad (23)$$

is the total probability of the decay from the level j to the metastable level i, and

$$w_{j,i}(j_1\ldots j_m)=(\Gamma_{jj_1}/2\Gamma_j)\ldots(\Gamma_{j_mi}/2\Gamma_{j_m}) \qquad (24)$$

is the probability of the decay proceeding via the channel $j\to j_1\ldots j_m\to i$.

5. NONLINEAR SUSCEPTIBILITY IN THE STANDING-WAVE FIELD

Consider a gas of $(q+1)$-level atoms in the field (2). Let the transition $q-1\to q$ be resonant to a standing wave and the remaining transitions resonant to the traveling waves $\mathscr{E}_q=2E_q\cos(k_qz)$, $\mathscr{E}_j=E_j\exp(ik_jz)$, where k_j is the wave vector of the jth wave and the z axis is the wave propagation direction. The susceptibility of the medium is defined as the coefficient of the proportionality of the polarization projection $P_q(z)$ on the standing-wave field

$$\chi=\int_0^{2\pi/k_q}\frac{dz}{(\pi/k_q)}\cos(k_qz)P_q(z)/E_q.$$

We obtain for the susceptibility

$$\chi=-|E_q|^{-2}\int_0^{2\pi/k_q}\frac{dz}{(\pi/k_q)}\int_{-\infty}^{\infty}dv\,\mathrm{Sp}(\rho A_{-q}),$$

where v is the projection of the atom velocity on the z axis. We take the relaxation into account by introducing into the left-hand sides of the equations for ρ_{ik} the terms $\gamma_{ik}\rho_{ik}$, where $\gamma_{ii}=\gamma_i$ is the rate of decay of the i-th metastable level, and $\gamma_{0q}=\gamma_{q0}=\gamma$. In a gas we have $d/dt=\partial/\partial t+v\partial/\partial z$. We transform to new variables

$$v\to k_qv,\quad z\to k_qz,\quad k_j\to k_j/k_q.$$

Let the level 0 be populated in the absence of a field. We seek the solution of (10) in the form

$$\rho_{q0}(z,t)=\exp(-i\nu t+i\eta z)G\rho_0^0\bar r(z),\quad \rho_{ii}(z,t)=\rho_0^0\rho_i(z),$$

where $\rho_0^0=NW_M(v)$, N is the gas density,

$$W_M(v)=(\pi^{1/2}k_qv_0)^{-1}\exp[-(v/k_qv_0)^2]$$

is a Maxwellian distribution function in velocity, v_0 is the thermal velocity, $G=-iG_1\ldots G_q(\Gamma_1\ldots\Gamma_{q-1})^{-1}$ (here and below $G_j=-d_{j,j-1}E_j$), and the parameter

$$\eta=\left|\sum_{j=1}^{q-1}k_j\right| \qquad (25)$$

is the algebraic sum of the wave vectors of the traveling waves in units of k_q. We then obtain

$$\left(v\frac{d}{dz}+\gamma-i(\nu-\eta v)\right)\bar r=2\cos z(\mathscr{L}\rho_0-\mathscr{L}^*\rho_q)$$
$$-\left(4\cos^2z\frac{|G_q|^2}{\Gamma_{q-1}}\mathscr{L}_q+\frac{|G_1|^2}{\Gamma_1}\mathscr{L}_0\right)\bar r,$$

$$\left(v\frac{d}{dz}+\gamma_0+\frac{2|G_1\mathscr{L}_0|^2}{\Gamma_1}\right)\rho_0=-4|G|^2\cos z\,\mathrm{Re}(\mathscr{L}\bar r)+\gamma_0,$$

$$\left(v\frac{d}{dz}+\gamma_q+\frac{8|G_q\mathscr{L}_q|^2}{\Gamma_{q-1}}\cos^2 z\right)\rho_q=4|G|^2\cos z\,\mathrm{Re}(\mathscr{L}^*\bar{r}),\tag{26}$$

where

$$\mathscr{L}=[(\delta_1+i)\ldots(\delta_{q-1}+i)]^{-1},$$

$$\mathscr{L}_0=(1+i\delta_1)^{-1},\quad \mathscr{L}_q=(1+i\delta)^{-1},$$

and $\delta_j=\Sigma_j/\Gamma_j$, $\delta=\Omega_q/\Gamma_{q-1}$. We shall be interested hereafter in the susceptibility saturation due to the nonlinear interaction with only the standing-wave field. Assuming the remaining fields to be weak and solving (26) by iterating over G, we obtain for $m=\rho_q/|\mathscr{L}G|^2, r=\bar{r}/\mathscr{L}$ the equations

$$\hat{\gamma}_s r=2\cos z,\quad \hat{\gamma}_q m=4\cos z\,\mathrm{Re}\,r,\tag{27}$$

where the differential operators are given by

$$\begin{aligned}\hat{\gamma}_s&=v\frac{d}{dz}+\gamma(1+2\varkappa\mathscr{L}_q\cos^2 z)-i(\nu-\eta v),\\ \hat{\gamma}_q&=v\frac{d}{dz}+\gamma_q(1+4\varkappa_q|\mathscr{L}_q|^2\cos^2 z),\end{aligned}\tag{28}$$

and the saturation parameters are $\varkappa=2|G_q|^2/(\Gamma_{q-1}\gamma)$, $\varkappa_q=2|G_q|^2/(\Gamma_{q-1}\gamma_q)$. It can be seen that besides the usual parameter $\varkappa_q$ that corresponds to saturation of the population of level q there appears a new parameter corresponding to saturation of the polarization of the transition $0\to q$.

From (20) we obtain for the susceptibility at $v_0\gg\gamma/k_q$

$$\chi=-i\frac{\alpha_0}{2\pi k_q}F(\nu),$$

where $\alpha_0=4\pi^{3/2}|G\mathscr{L}/G_q|^2N|d_{q,q-1}|^2/v_0$,

$$F(\nu)=\int_0^\infty\frac{dv}{\pi}(\Phi(v)+\text{the same with the substitution } \eta\to-\eta),\tag{29}$$

$$\Phi(v)=\langle\gamma\varkappa\mathscr{L}_q^*m\cos^2 z-r\cos z\rangle,\tag{30}$$

and the averaging is along z. The absorption coefficient α and the refractive index n are equal to

$$\alpha=-2\alpha_0\,\mathrm{Re}\,F,\quad n=1+(\alpha_0/k_q)\,\mathrm{Im}\,F.\tag{31}$$

Being interested only in the nonlinear part of χ, we can make the substitution $r\to r-r_l$, where

$$r_l=2\hat{\gamma}_l^{-1}\cos z,\quad \hat{\gamma}_l=v\frac{d}{dz}+\gamma-i(\nu-\eta v),\tag{32}$$

since r_l corresponds to the linear susceptibility of the gas.

We confine ourselves here to the case of a weak field, $\varkappa\ll1$. Solving (27) by iterating over $\varkappa$, we obtain ultimately

$$F(\nu)=\frac{2\varkappa_q}{|1-\eta^2|}\left[\theta(1-\eta)\left(\mathscr{L}_q^*+\mathscr{L}_q\frac{(1-\eta^2)}{4}\frac{\gamma_q}{\gamma_R-i\nu}\right)+\eta\theta(\eta-1)\mathscr{L}_q^*\left(1+\frac{\eta^2-1}{4\eta}\frac{\gamma_q\gamma_R}{\gamma_R^2+\nu^2}\right)\right],\tag{33}$$

where

$$\gamma_R=\begin{cases}\gamma, & \eta<1,\\ \gamma+\dfrac{\eta-1}{2}\gamma_q, & \eta>1.\end{cases}$$

6. STRONG FIELD

Let now $\varkappa\sim\varkappa_q\sim1$. We assume for simplicity that $\Omega_q=0$. It is more convenient to calculate α and n separately. Direct calculation shows that if

$$\Phi(v)=2\langle\cos z(\hat{\gamma}^{-1}-\hat{\gamma}_l^{-1})\cos z\rangle,\tag{34}$$

where

$$\hat{\gamma}^{-1}=\int_{-\infty}^{z}\frac{dz_1}{v}\exp[-(\gamma(1+\varkappa)-i(\nu-\eta v))(z-z_1)/v],$$

is substituted in (29), we obtain an expression identically equal to zero. Substituting in (30) the solution (27) and adding to $\Phi(v)$ [Eq. (34)], with allowance for the substitution $r\to r-r_l$, we obtain after elementary integration with respect to z and v the nonlinear increment $\Delta n=(\alpha_0/k_q)Q(\nu)$ to the refractive index, with

$$Q(\nu)=\frac{2}{\pi}\mathrm{Im}\int_0^\infty dr\cos(\eta r)\{[1-(1-(p\sin r/r)^2)^{1/2}]/(p\sin r/r)+\cos r\ln[(1+(1-(p\sin r/r)^2)^{1/2})/2]\},\tag{35}$$

where $p=\gamma\varkappa/(\gamma(1+\varkappa)-i\nu)$.

We now calculate α. At $\Omega_q=0$, $\mathrm{Re}\,\Phi(v)$ decreases quite rapidly as a function of v, so that our equations suffice for the calculation of both the linear and nonlinear parts of α. From (27) we get

$$\mathrm{Re}\,\Phi(v)=-\langle\cos z\,\mathrm{Re}\,r\rangle+4\gamma\varkappa\int_{-L}^{L}\frac{dz}{2L}\int_{-L}^{z}\frac{dz_1}{v}\cos^2 z\cos z_1 A(z,z_1)\,\mathrm{Re}\,r(z_1),\tag{36}$$

where L is the normalization length along z, and

$$A(z,z_1)=\exp\left[-\gamma_q\int_{z_1}^{z}\frac{dz'}{v}(1+4\varkappa_q\cos^2 z')\right].$$

Changing the sequence of the integration in the second term of (36) and separating the term proportional to $\partial A(z,z_1)/\partial z$, we verify that it cancels completely the first term of (36). In the upshot we get

$$\mathrm{Re}\,\Phi(v)=-\gamma_q\langle\hat{\gamma}_q^{-1}\cos z\,\mathrm{Re}\,r\rangle.\tag{37}$$

Substituting this expression in (29) we get ultimately

$$\begin{aligned}\mathrm{Re}\,F(\nu)&=-\frac{\theta(1-\eta)+\eta\theta(\eta-1)}{(1+2\varkappa_q)|1-\eta^2|}+J(\nu),\\ J(\nu)&=\frac{2}{\pi\varkappa}\mathrm{Re}\int_0^1 dx\,p(x)\varphi(x),\\ \varphi(x)&=\int_0^\infty dr\cos(\eta rx)[(1-(p(x)s)^2)^{-1/2}-1]\end{aligned}\tag{38}$$

$$\left\{\frac{\cos[\varphi+r(2-x)]}{p(x)s}-\cos(rx)\right\},$$

$$s\exp(i\varphi)=\frac{1}{ir}\left[1-\frac{1}{2}(\exp(2ir(x-1))+\exp(-2ir))\right],$$

$$p(x)=(\alpha x+\beta)^{-1}, \tag{39}$$

$$\alpha=(\gamma(1-\varkappa)-\gamma_q-i\nu)/\gamma\varkappa, \quad \beta=2+\varkappa_q^{-1}.$$

The first term in (38) yields the background of the resonance, and the function $J(\nu)$ the resonance proper. Characteristically, the saturation parameters in (35) and (39) enter in combinations

$$\varkappa/(a+\varkappa), \tag{40}$$

that remain finite as $\varkappa\to\infty$. Expansions is terms of these quantities should converge more rapidly than expansions in powers of $\varkappa$. They can be obtained by carrying out the integration along a straight line C shifted by an infinitely small amount into the lower half of the complex r plane. Expanding the integrands in (35) and (36) in powers of p and $p(x)$ we obtain the integral

$$\theta_m(a)=\frac{m!}{2\pi i^{m+1}}\int\limits_C\frac{dr}{r^{m+1}}\exp(ira).$$

Closing its contour C in the upper (at $a>0$) or lower (at $a<0$) half-plane, we get $\theta_m(a)=a^m\theta(a)$. This equation suffices to obtain any term of the expansion in the parameter (40), and following the expansion in (39) the integration with respect to $\varkappa$ also becomes elementary. The lengthy final formulas were given by us in Ref. 16.

We have calculated the functions Q and J from Eqs. (35) and (39). The results of the calculation of the resonance in the refractive index are given in Ref. 18 (see also Ref. 16). We present below the results for the absorption resonance. We characterize it by the amplitude $A=J_{\max}-J_{\min}$ and by a half-width which we define as equal to the maximum root of the equation

$$J(\nu)={}^1/_2(J_{max}+J_{min}). \tag{41}$$

Figures 1 and 2 show the dependences of A and γ_R on $\varkappa$. The jumps of the $\gamma_R(\varkappa)$ plots are due to the fact that the deformation of the contour of $J(\nu)$ changes the number of the roots of Eq. (41). The dashed lines in the figures show approximate dependences of the resonance parameters that appear when (39) is expanded in $p(x)$ up to the 10th term.

7. ASYMPTOTIC CASE

The function $J(\nu)$ at $\varkappa\to\infty$ is shown in Fig. 3. We are interested here in small corrections to this contour. It can be seen from (35) and (39) that at $\nu=\gamma=0$ the integrands contain branch points at $r=0$. Consequently Q and J are nonanalytic functions of ν near the line center. Let us ascertain the character of this nonanalyticity. We consider absorption resonance. At $\nu/\gamma\sim 1\ll\varkappa$ we have

$$p(x)=(1-\delta)/(1+y), \quad \delta=[\gamma_q+(\gamma-\gamma_q-i\nu)x]/[\gamma\varkappa(1+y)], \tag{42}$$

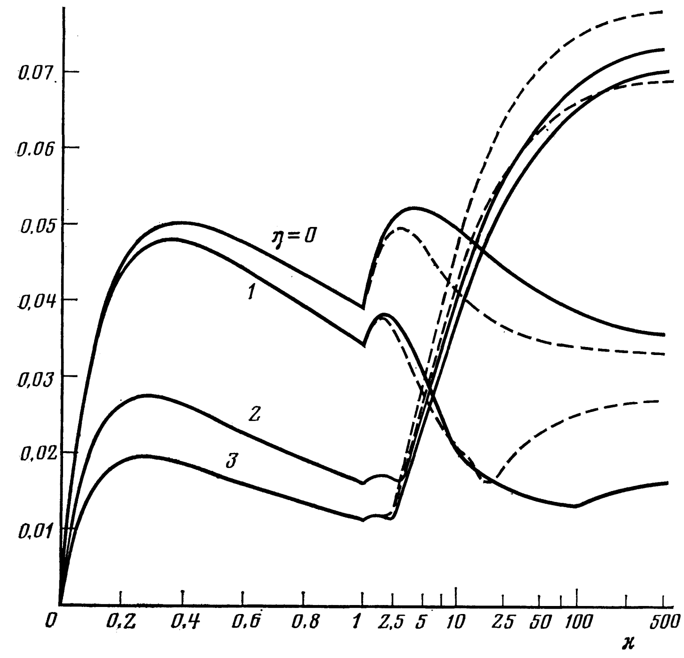

FIG. 1. Exact (solid curve) and approximate (dashed) field dependences of the resonance amplitude $A\ [\vartheta(1-\varkappa)+\varkappa\vartheta(\varkappa-1)]$ in absorption at $\gamma=\gamma_q$ for different values of the parameter η.

where $y=1-x$. We divide the region of integration with respect to r in two, $r<a$ and $r>a$, where $\delta\ll a\ll 1$, and denote their contributions to $\varphi(x)$ by φ_1 and φ_2. At small r we have $s^2=(1+y)^2(1-fr^2)$, and the factor in the curly brackets in (39) is equal to $\delta+r^2f_1$, where

$$f=[{}^4/_3(1+y)(1+y^3)-(1+y^2)^2]/(1+y)^2,$$

$$f_1=[{}^2/_3(1+y)(1+y^3)-4y^2]/(1+y)^2.$$

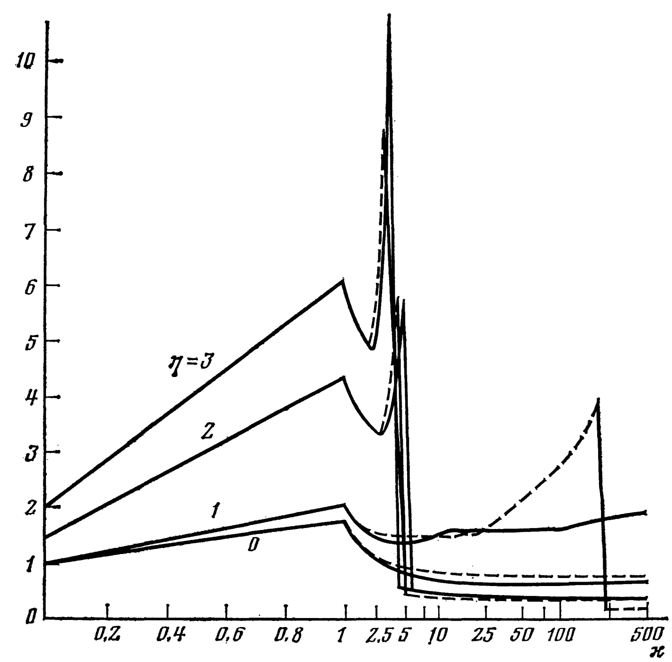

FIG. 2. The same as Fig. 1 but for the absorption resonance half-width (the ordinates are $\gamma_R\ [\vartheta(1-\varkappa)+\varkappa^{-1}\vartheta(\varkappa-1)]$).

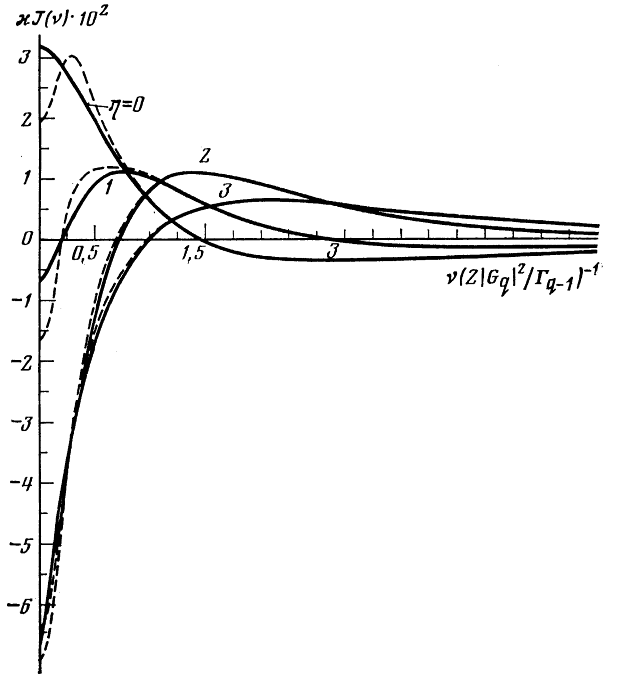

FIG. 3. Exact (solid curve) and approximate (dashed) shape of resonant increment to the absorption coefficient as $\varkappa \to \infty$ for different values of the parameter η.

We then obtain for φ_1

$$\varphi_1 = -{}^1/{}_3 a^3 f_1 + {}^1/{}_2 f_1 f^{-1/2} a^2 + \delta (f - f_1) f^{-3/2} \ln a + {}^1/{}_2 \delta f^{-3/2} [(f - f_1) \ln (2f/\delta) + f_1]. \tag{43}$$

When integrating over the region $r > a$ we can expand the integrand in powers of δ. Then φ_2 acquires terms that cancel the first three terms of (43). We shall not present here the awkward formulas for the remaining terms of φ_2 (see Ref. 16), the more so since they yield no additional dependence of J on ν; we indicate only that they can be completely neglected at $\ln \varkappa \gg 1$. We then obtain from (43) for the increment $\delta J(\nu)$ to the limiting value of $J(0)$ from (39)

$$\delta J(\nu) = \frac{\ln \varkappa}{\pi \varkappa \varkappa} \int_0^1 dy [1 - y(1 - \gamma/\gamma_q)] (f - f_1) f^{-3/2} (1 + y)^{-2} + \frac{1}{\pi \varkappa \varkappa} \int_0^1 dy (f - f_1) f^{-3/2} (1 + y)^{-1} \left[\delta_2 \operatorname{arctg}(\delta_2/\delta_1) - \frac{1}{2} \delta_1 \ln (1 + \delta_2^2/\delta_1^2)\right], \tag{44}$$

where $\delta_1 + i\delta_2 = \varkappa\delta$. Differentiating (44), we see that in the first derivative of the absorption resonance there appears a line free of field broadening

$$\frac{dJ}{d\nu} = \frac{1}{\pi \gamma \varkappa \varkappa_q} \int_0^1 dy \frac{(1 - y)(f - f_1)}{(1 + y)^2 f^{3/2}} \operatorname{arctg} \left(\frac{\nu (1 - y)}{\gamma + y(\gamma_q - \gamma)} \right). \tag{45}$$

The evolution of $dJ/d\nu$ with increasing $\varkappa$ in the region $\nu \sim \gamma$ is shown in Fig. 4.

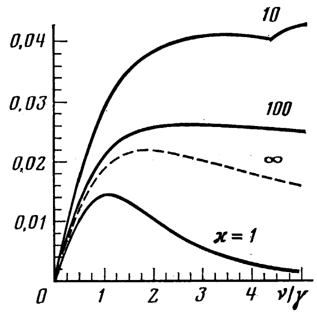

FIG. 4. Evolution, with increasing field intensity, of the derivative of the resonant increment to the absorption coefficient at $\gamma = \gamma_q$ and $\eta = 0$. The dashed line is a plot of (45); the ordinates are $-\varkappa^2 \gamma dJ(\nu)/d\nu$.

The line $Q(\nu)$ is treated analogously. The integrand should be expanded here in powers of $(1 - (p \sin r/r)^2)^{1/2}$ to the cubic term, which is the one responsible for the nonanalytic increments. A resonance free of field broadening appears[16] only in the third derivative and has a Lorentz shape

$$\frac{d^3 Q}{d\nu^3} = -\frac{3^{1/2}}{\pi \gamma \varkappa^2} (\gamma^2 + \nu^2)^{-1}. \tag{46}$$

8. DISCUSSION

The foregoing derivation of the equations for the GTS differs from that in Refs. 14 and 15. We used an S matrix in place of an expression for $\bar{\sigma}$ in the form of multiple commutators. In q-th order perturbation theory we obtained for $\bar{\sigma}$ only $q + 1$ terms as against 2^q in Refs. 14 and 15. That the effective Hamiltonian of the nonresonant situation is Hermitain can be established without resorting to the explicit form of U, and follows from the unitarity of the S matrix. Indeed, retaining in the vanishing q-th term of the expansion of SS^+ in terms of V only the terms linear in T, we obtain $S_q = -S_q^+$, which verifies that the Hamiltonian (11) is Hermitian. At $\Gamma \neq 0$ the matrix is not unitary and U is not Hermitian. It follows therefore that the probability of finding the atom on metastable levels varies with time, $\mathrm{Sp}\rho \neq \mathrm{const}$. Yet it is obvious that for a conservative GTS, in which the end products of the decay are only the level 0 and q, the probability should be conserved. Even this fact alone points to a fundamental role of the processes of real population of the intermediate levels. That the equations (22) which result from allowance for these processes lead to $\mathrm{Sp}\rho = \mathrm{const}$ can be verified by taking it into account that $A_{-1} + A_q = U$ and that $w_{j1} + w_{jq} = 1$ for a conservative GTS.

Our approach to the calculation of the response of the system is also different. The use of the set of constants λ_j made it possible to establish a direct operator connection between D, U, and S. At $\Gamma = 0$ (in the nonresonant situation)

$$D_j = -\mathscr{E}_{-j}^{-1} \partial U / \partial \lambda_{-j}. \tag{47}$$

It can be seen that there is no need here to divide[14,15] D into resonant and nonresonant parts, and that the proportionality of the off-diagonal elements of D to the positive- or negative-frequency parts of U follows from the vanishing of the derivative, with respect to λ, of the terms with incorrect frequency dependence. We note that the expressions (13) for $U_{i,k}$ are obtained from the formulas for U at $\Gamma = 0$ if only the resonant terms are retained and if an imaginary increment $-i\Gamma_l$ to the energy of the final level l is added to each of the denominators. This, however, is not enough to obtain correct expressions for D. In fact, in the opposite case Eq. (47) would remain valid at $\Gamma \neq 0$, but it follows from (19) that this is not the case.

Equations for the incomplete density matrix were already used earlier in the theory of nonlinear resonances. Thus, to calculate the coherent corrections to the Lamb dip, Baklanov and Chebotaev[11] obtained for the population difference and equation that can be intrepreted as an equation for the density matrix of a structureless particle with a non-Hermitain Hamiltonian that is quadratic in the field of the standing wave. In Ref. 17 were obtained GTS equations for the case of a two-photon resonance. In the absence of the Doppler effect (in the radio band or for particles captured in a trap), the GTS method can be used to calculate the total susceptibility. In our case it can be used only to calculate the resonant increment to χ. The nonresonant background is usually calculated by considering the complete system of equations, and in this case the rate approximation[18] is applicable.

An important question in the theory of nonlinear resonances is that of the field dependence of the parameters that describe the resonances. This dependence is calculated by using perturbation theory, the rate approximation and the corrections to it, and in our case one can use expansions in terms of the parameter (40). The availability of an exact solution permits an estimate of the ranges of validity of these approximations. It is found, for example, that the field dependences obtained by perturbation theory (see Ref. 16) are valid, accurate to 10%, for the amplitude and width of the absorption resonance up to $\varkappa$ values 0.05 and 0.8, respectively. This example shows that the perturbation-theory calculation frequently used in applications must be approached with some caution, and should be used only when the range of their validity has been determined. It can be seen from Figs. 1 and 2 that expansions in terms of the parameter (40), with only the first 10 terms retained, yield the same accuracy up to $\varkappa = 2.5$ for absorption. It is seen from Fig. 3 that as $\varkappa \to \infty$ the approximate solution agrees wtih the exact one at sufficiently large ν. Thus, an accuracy to better than 10% is reached at $\nu > 1.15\,\gamma\varkappa$. We note that it is convenient to use the expansions also outside this region, since a possibility arises of lowering the upper limit of integration in (35) and (39) by 1–2 orders of magnitude.

At small $\varkappa$ the absorption line is Lorentzian. It can be seen from Fig. 3 that the line becomes deformed when $\varkappa$ is increased. It is known that in the case of homogeneous saturation there is no deformation, therefore we attribute the latter to spatial modulation of the medium.

As $\varkappa \to \infty$ the amplitude of the resonance tends to a constant value for the refractive index and decreases in proportion to $\varkappa^{-1}$ for absorption. The line width increase in proportion to $\varkappa$:$\gamma_R \sim \gamma\varkappa = 2|G_q|^2/\Gamma_{q-1}$ and ceases to depend on γ. It can thus be concluded that at large $\varkappa$ the line shape is a universal function of the standing-wave intensity and is independent of the mechanism of the relaxation of the metastable levels.

At the same time, singularities free of field broadening are preserved in the line. They lead to the appearance of lines having a width on the order of γ in the higher derivatives of χ with respect to ν. We attribute this effect to the anomalous growth of the spatial modulation of the gas polarization. To describe it we must sum exactly the contributions from all the higher spatial harmonics of the density matrix. It is interesting to note that the same effect takes place also for the Lamb dip at $\gamma \ll \Gamma$. The narrow line appears here in the third derivative of the absorption resonance. From the equations of Ref. 11 we obtain for it

$$\frac{d^3\alpha}{d\Omega^3} = \frac{3^{1/2}\gamma}{\pi\Gamma\varkappa^2}\alpha_0{}' \frac{\Omega(3\Gamma^2+\Omega^2)}{[\Gamma(\Gamma^2+\Omega^2)]^2},$$

where α_0' is the unsaturated absorption coefficient. The lack of equations in quadratures leaves open the question of the presence of this effect in one-photon nonlinear spectroscopy at $\gamma \sim \Gamma$. We note that this effect can substantially distort the field dependence of the shift of the nonlinear resonance, since the shift is usually sensitive to the higher derivatives of the resonance. No narrow line is produced on differentiation with respect to $\varkappa$, i.e., at $\varkappa \gg 1$ the situation differs qualitatively from the weak-field case, where differentiation with respect to frequency and intensity leads to identical effects (cf. the results of Refs. 19 and 20).

Suitable objects for the experimental observation of the multiphoton line are atomic gases. I cited in Ref. 16 examples of multilevel schemes of alkali and alkaline-earth atoms in which the transition frequencies land in the tuning range of cw lasers. In these lasers a narrow resonance can already be observed and used at present.

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[1]The alternate approach[7,8] based on the Stark effect is hardly suitable for precision spectroscopy, since it calls for high-power radiation of relative stability on the order of $\gamma/\omega_D \ll 1$.

[2]It is known[10] that the rate approximation can also be used at $\gamma \sim \Gamma$ if the splitting of the Lamb dip by the recoil is large. Its use in other cases is not justified.

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