Contribution to the theory of nonlinear power resonances in gas lasers

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The effect of collisions on the shape of nonlinear power resonances in gas lasers is investigated. The resonance shape is found to be very sensitive to fine details of the collision mechanism, which are usually unobservable in the linear absorption and emission spectra. The angular dependence of the scattering amplitude, the relation between elastic and inelastic scattering channels, etc. are found to be important. It is shown that under certain conditions the resonance width and shift become nonlinear functions of the perturbing atom density.

1. INTRODUCTION

In many problems of quantum electronics (stabilization of the frequency of gas lasers, high-resolution laser spectroscopy, etc.), the structure of narrow power resonances, of the so-called "Lamb-dip" type[1], which occur when the frequency of gas lasers is scanned, is of great interest. The shape and position of these resonances are sensitive to the fine details of the interaction mechanism of the atoms and molecules with the surrounding particles[2-5]. Many of the effects that are produced in this case are not observed in practice in ordinary linear absorption and emission spectra, for in this case the role of the collisions is masked by the broad Doppler contour of the line. In nonlinear resonances, the Doppler broadening is practically eliminated, so that special requirements must be satisfied to describe the collisions. The phenomenological approach extensively used in many problems of quantum electronics and nonlinear optics, based on introducing relaxation terms or model collision integrals into the equation for the density matrix of the atom, is too crude in the present case.

In this paper we describe nonlinear power resonances of gas lasers by using an equation derived in[6] for the density matrix. This equation yields for the atomic collisions the most complete description compatible with the gas-kinetic approach, i.e., compatible with the assumption that the perturbations produced by the particles that surround the atom reduce to time-separated collisions[1)]. In the equation we consider in a unified quantum fashion both the motion of the atomic electron and the motion of the atom as a unit; the collision integral is expressed in terms of exact scattering amplitudes with allowance for both elastic and inelastic channels. This approach enables us to investigate the change in the form of the nonlinear resonances in the entire pressure range of interest for applications.

It turns out that even within the framework of the impact approximation, i.e., at relatively low pressure, the width and shift of the resonances become, under definite conditions, nonlinear functions of the pressure, a fact that can be of primary importance for the stabilization of gas-laser frequencies. Nonlinear dependences of the width and shift of the resonance on the pressure were observed recently in experiments and qualitatively explained by Bagaev, Baklanov, and Chebotaev[3,4].

2. INTERACTION OF THE ATOM WITH A FIELD OF A MONOCHROMATIC STANDING WAVE

We consider an atomic or molecular gas situated in a field of a standing wave

$$\mathbf{E}(\mathbf{R}, t) = \mathbf{E}_0 \cos(\omega t - \mathbf{pR}), \quad p = \omega / c,$$

We describe the atom by means of the following equation for the density matrix ρ (see[6]):

$$\frac{d\rho}{dt} = \frac{i}{\hbar}[H_0, \rho] + \frac{i}{\hbar}\mathbf{E}_0[\mathbf{d}\cos(\omega t - \mathbf{pR}_a), \rho] + \left(\frac{d\rho}{dt}\right)^{\text{col}}. \quad (1)$$

Here H_0 is the Hamiltonian of the free atom, which includes the kinetic energy of the motion of the atom as a whole, d is the dipole moment of the atom, R_a is the coordinate of the mass center of the atom, and $(d\rho/dt)^{col}$ is a term describing the collision of the atom with the surrounding particles. Concrete expressions for $(d\rho/dt)^{col}$ will be presented below.

We shall calculate later on the average absorption power (or emission power) $P(\omega)$ of the atom [erg/sec] at the field frequency. This quantity is determined by the expression

$$P(\omega) = \frac{1}{2}\mathbf{E}_0 \operatorname{Sp}[\dot{\mathbf{d}} e^{i\mathbf{pR}_a}\rho(\omega) + \dot{\mathbf{d}}^* e^{-i\mathbf{pR}_a}\rho^*(\omega)].$$

Calculating the trace in the mk representation, where m are the quantum numbers of the stationary states of the atom and k is the wave vector of the motion of the atom as a whole, and recognizing that

$$(\dot{\mathbf{d}} e^{i\mathbf{pR}_a})_{m\mathbf{k}, n\mathbf{k}'} \propto \dot{\mathbf{d}}_{mn}\delta(-\mathbf{k} + \mathbf{p} + \mathbf{k}'),$$

we obtain

$$P(\omega) = 2\hbar\omega_0 \int d\mathbf{k}\, u(\mathbf{k}),$$

$$u(\mathbf{k}) = \operatorname{Re}\lambda^*[\rho_{mn}(\omega, \mathbf{k}, \mathbf{k}+\mathbf{p}) + \rho_{mn}(\omega, \mathbf{k}, \mathbf{k}-\mathbf{p})],$$

where $\lambda = (i/2\hbar)\mathbf{E}_0 \cdot \mathbf{d}_{mn}$; $\rho_{mn}(\omega, \mathbf{k}, \mathbf{k}')$ is the Fourier component, and $\rho_{mn,nk'}(t) \equiv \rho_{mn}(t, \mathbf{k}, \mathbf{k}')$.

We shall consider throughout the spectral characteristics of the medium at frequencies ω close to the frequency ω_0 of the $m \to n$ atomic transition. For problems of this type we confine ourselves to the two-level approximation and discard from the right-hand side of (1) the terms that lead to oscillations of solutions with double the frequency. As a result we obtain from (1)

$$\dot{\rho}_{mn}(\mathbf{k}, \mathbf{k}') - i\left[\omega_0 + \frac{\hbar}{2m_a}(k^2 - k'^2)\right]\rho_{mn}(\mathbf{k}, \mathbf{k}') - \left(\frac{d\rho}{dt}\right)^{\text{col}}_{\mathbf{mk}, n\mathbf{k}'} \quad (2)$$
$$= \lambda e^{i\omega t}[\rho_{nn}(\mathbf{k}+\mathbf{p}, \mathbf{k}') + \rho_{nn}(\mathbf{k}-\mathbf{p}, \mathbf{k}') - \rho_{mm}(\mathbf{k}, \mathbf{k}', -\mathbf{p}) - \rho_{mm}(\mathbf{k}, \mathbf{k}'+\mathbf{p})],$$

$$\dot{\rho}_{mn}(\mathbf{k},\mathbf{k}')-i\frac{\hbar}{2m_a}(k^2-k'^2)\rho_{mn}(\mathbf{k},\mathbf{k}')-\left(\frac{d\rho}{dt}\right)^{\mathrm{col}}_{mk,mk'}$$
$$=\lambda e^{i\omega t}[\rho_{nm}(\mathbf{k}+\mathbf{p},\mathbf{k}')+\rho_{nn}(\mathbf{k}-\mathbf{p},\mathbf{k}')]$$
$$+\lambda^* e^{-i\omega t}[\rho_{mn}(\mathbf{k},\mathbf{k}'+\mathbf{p})+\rho_{mn}(\mathbf{k},\mathbf{k}'-\mathbf{p})], \quad (3)$$

$$\dot{\rho}_{nn}(\mathbf{k},\mathbf{k}')-i\frac{\hbar}{2m_a}(k^2-k'^2)\rho_{nn}(\mathbf{k},\mathbf{k}')-\left(\frac{d\rho}{dt}\right)^{\mathrm{col}}_{nk,nk'}$$
$$=-\lambda e^{i\omega t}[\rho_{nm}(\mathbf{k},\mathbf{k}'-\mathbf{p})+\rho_{nm}(\mathbf{k},\mathbf{k}'+\mathbf{p})]-$$
$$-\lambda^* e^{-i\omega t}[\rho_{mn}(\mathbf{k}-\mathbf{p},\mathbf{k}')+\rho_{mn}(\mathbf{k}+\mathbf{p},\mathbf{k}')], \quad (4)$$

with $\rho_{nm}(\mathbf{k},\mathbf{k}')=\rho^*_{mn}(\mathbf{k}',\mathbf{k})$.

The system (2)—(4) enables us to find the characteristics of the medium that are both linear and nonlinear in the field, with allowance for the motion of the atoms and for collisions. It is seen from this system that $\rho_{mn}(\mathbf{k},\mathbf{k}',t)=\rho_{mn}(\omega,\mathbf{k},\mathbf{k}')e^{i\omega t}$, and $\rho_{ii}(t,\mathbf{k},\mathbf{k}')=\rho_{ii}(\mathbf{k},\mathbf{k}')$ does not depend on the time. The function $P(\omega)$ is expressed in terms of the matrix elements $\rho_{mn}(\omega,\mathbf{k},\mathbf{k}\pm\mathbf{p})$, the determination of which calls for solution of Eq. (2)—(4). When $\rho_{mn}(\omega,\mathbf{k},\mathbf{k}')e^{i\omega t}$ is substituted in the initial system (2)—(4), we obtain a chain of equations for the quantities

$$\rho_{mn}(\omega,\mathbf{k}+l\mathbf{p},\mathbf{k}+l'\mathbf{p}),\quad l'-l=2j+1,$$
$$\rho_{mm}(\mathbf{k}+l\mathbf{p},\mathbf{k}+l'\mathbf{p}),\quad \rho_{nn}(\mathbf{k}+l\mathbf{p},\mathbf{k}+l'\mathbf{p}),\quad l'-l=2j;$$
$$j=0,\pm1,\pm2\ldots.$$

This system is similar in its structure to the equations obtained by Feldman and Feld[9] in which, however, the motion of the center of mass of the atom was treated classically, and the collisions were described by introducing phenomenological relaxation constants.

We shall use in what follows the following simplifications: a) We confine ourselves, as is customary, to third order of perturbation theory in the field, i.e., we seek solutions with accuracy to terms of order λ^3 inclusive. b) Bearing in mind the most typical experimental conditions, when the homogeneous line width is much smaller than the Doppler width, we disregard the so-called "spatial burning out of the atoms" when we consider third-order perturbation theory in the field, i.e., we neglect the matrix elements ρ_{ii} at $l\neq l'$. c) Since $p\ll k$, we assume that in the absence of a field the distribution of the atoms with respect to the wave vectors $W(\mathbf{k})$ remains practically unchanged when the argument is shifted by an amount p, namely, $W(\mathbf{k}+\mathbf{p})\approx W(\mathbf{k})$. We can therefore make the substitution

$$\rho_{mn}(\mathbf{k}\pm\mathbf{p},\mathbf{k}\pm2\mathbf{p})\approx\rho_{mn}(\mathbf{k},\mathbf{k}\pm p).$$

We shall henceforth omit the index ω of the matrix elements $\rho_{mn}(\omega,\mathbf{k},\mathbf{k}')$. We introduce also the notation

$$\rho_{ii}(\mathbf{k},\mathbf{k})=z_iW(\mathbf{k})+\rho_i(\mathbf{k}),\quad z=z_m-z_n,$$

where z_i is the population of the state i.

Using the approximations b) and c), we can obtain from (2)—(4) the following system of equations:

$$i\left(\omega-\omega_0\pm\frac{\hbar}{m_a}k_xp\right)\rho_{mn}(\mathbf{k},\mathbf{k}\pm\mathbf{p})-\left(\frac{dp}{dt}\right)^{\mathrm{col}}_{mk,nk\pm p}$$
$$=\frac{\lambda}{2}[zW(\mathbf{k})+\rho_n(\mathbf{k})-\rho_m(\mathbf{k})], \quad (5)$$

$$\left[\frac{d}{dt}\rho_n(\mathbf{k})\right]^{\mathrm{col}}=\mathrm{Re}\,\lambda^*[\rho_{mn}(\mathbf{k},\mathbf{k}+\mathbf{p})+\rho_{mn}(\mathbf{k},\mathbf{k}-\mathbf{p})], \quad (6)$$

$$-\left[\frac{d}{dt}\rho_m(\mathbf{k})\right]^{\mathrm{col}}=\mathrm{Re}\,\lambda^*[\rho_{mn}(\mathbf{k},\mathbf{k}+\mathbf{p})+\rho_{mn}(\mathbf{k},\mathbf{k}-\mathbf{p})],$$

$$\left[\frac{d}{dt}\rho_i(\mathbf{k})\right]^{\mathrm{col}}=\left(\frac{d\rho}{dt}\right)^{\mathrm{col}}_{ik,ik}; \quad (7)$$

The x axis is chosen along the vector p.

3. INFLUENCE OF COLLISIONS

We use first a simplified phenomenological description of the collisions, putting

$$\left(\frac{d\rho}{dt}\right)^{\mathrm{col}}_{mk,nk\pm p}=-\left(\frac{\Gamma}{2}+i\Delta\right)\rho_{mn}(\mathbf{k},\mathbf{k}\pm\mathbf{p}); \quad (8)$$

$$\left[\frac{d}{dt}\rho_m(\mathbf{k})\right]^{\mathrm{col}}=\beta_m\rho_m(\mathbf{k}),\quad \left[\frac{d}{dt}\rho_n(\mathbf{k})\right]^{\mathrm{col}}=\beta_n\rho_n(\mathbf{k}), \quad (9)$$

where Γ and Δ are the width and shift of the spectral line in the impact theory of the broadening of spectral lines; β_m and β_n are the relaxation constants, i.e., the reciprocal lifetimes of the states m and n. In (9) we make use of the fact that the equilibrium value $\rho_{ii}=z_iW(\mathbf{k})$ causes the collision term to vanish.

Solving the system (5)—(7) in third order perturbation theory in the field, i.e., in the parameter λ, we can easily obtain the following expression for the quantity $P(\omega)$:

$$P(\omega)=2\pi\hbar\omega_0 z|\lambda|^2I(\omega)[1-|\lambda|^2C(\omega)]. \quad (10)$$

The first term of this expression, which is proportional to $|\lambda|^2$, gives the well-known contour of the linear absorption. The function $I(\omega)$ is the convolution of a Lorenz contour with width Γ and shift Δ of the maximum, on the one hand, with a Doppler contour, on the other. In the case when Γ and Δ are small in comparison with the Doppler width $\Delta\omega_D$, the function $C(\omega)$ describes a narrow resonance of width Γ and shift Δ against the background $I(\omega)$:

$$C(\omega)=\left(\frac{1}{\beta_n}+\frac{1}{\beta_m}\right)\frac{1}{\Gamma}\left[1+\frac{\Gamma^2/4}{(\omega-\omega_0+\Delta)^2+\Gamma^2/4}\right]. \quad (11)$$

Resonances of this type were obtained in a number of papers (see, for example,[1]).

We proceed now to a more complete description of the collisions, using for the collision term $(d\rho/dt)^{\mathrm{col}}$ the expression obtained in[6]:

$$\left(\frac{d\rho}{dt}\right)^{\mathrm{col}}_{mk,nk\pm p}=-\left(\frac{\Gamma}{2}+i\Delta+\nu_{mn}\right)\rho_{mn}(\mathbf{k},\mathbf{k}\pm\mathbf{p})$$
$$+\int A_{mn}(\mathbf{k},\mathbf{k}')\rho_{mn}(\mathbf{k}',\mathbf{k}'\pm\mathbf{p})\,d\mathbf{k}', \quad (12)$$

$$\left[\frac{d}{dt}\rho_i(\mathbf{k})\right]^{\mathrm{col}}=-(\beta_i+\nu_i)\rho_i(\mathbf{k})+\int A_{ii}(\mathbf{k},\mathbf{k}')\rho_i(\mathbf{k}')\,d\mathbf{k}'. \quad (13)$$

Here

$$\frac{\Gamma}{2}+i\Delta=N\frac{\hbar}{\mu}\langle q(\sigma'+i\sigma'')\rangle$$
$$=N\frac{\hbar}{\mu}\int dq\,qW_p\left(\frac{\mu_p\mathbf{k}-\mathbf{q}}{\mu_a}\right)[\sigma'(q)+i\sigma''(q)], \quad (14)$$

where σ' and σ'' are the cross sections of the width and shift of the impact broadening theory; N is the concentration of the perturbing particles:

$$\mu=\frac{m_am_p}{m_a+m_p},\quad \mu_a=\frac{m_a}{m_a+m_p},\quad \mu_p=\frac{m_p}{m_a+m_p};$$

m_a and m_p are the masses of the atom and of the perturbing particle. The angle brackets in (14) denote the operation of averaging over the velocities of the perturbing particles, and W_p is the distribution function of the perturbing particles with respect to the wave vectors. Further,

$$\beta_i=N\frac{\hbar}{\mu}\langle q\sigma_i^{\mathrm{in}}\rangle,\quad \nu_i=N\frac{\hbar}{\mu}\langle q\sigma_i^{\mathrm{el}}\rangle,\quad \nu_{mn}=N\frac{\hbar}{\mu}\langle q\sigma_{mn}\rangle,$$

where σ_i^{in} and σ_i^{el} are the inelastic and elastic scattering cross sections for the state i. The cross section σ_{mn} plays the role of inelastic scattering for the off-diagonal matrix element; it differs from the usual

elastic-scattering cross section σ_m^{el} in that the square of the modulus of the scattering amplitude $|f_m|^2$ is replaced by $f_m f_n^*$, while the angle brackets denote again averaging analogous to (14). The integrand $A_{ij}(\mathbf{k}, \mathbf{k}')$ is connected with the amplitudes f_i and f_j for the scattering of the atom in the states i and j in the following manner:

$$A_{ij}(\mathbf{k}, \mathbf{k}') = N\frac{\hbar}{\mu}\int d\mathbf{q}\, W_p\left(\frac{\mathbf{k}-\mu_a\mathbf{k}'-\mathbf{q}}{\mu_a}\right)\delta(|\mathbf{k}'-\mathbf{k}+\mathbf{q}|-q) \times\frac{1}{q} f_i^*(\mathbf{k}'-\mathbf{k}+\mathbf{q}, \mathbf{q}) f_j(\mathbf{k}'-\mathbf{k}+\mathbf{q}, \mathbf{q}). \quad (15)$$

It will be convenient in what follows to write down expressions for σ', σ'', σ_{mn}, and σ_i^{in} in the form of sums over the partial waves l:

$$\sigma'(q) + i\sigma''(q) = \frac{\pi}{q^2}\sum_l (2l+1)[1 - S_n^l S_m^{l*}], \quad (16)$$

$$\sigma_{mn}(q) = \frac{\pi}{q^2}\sum_l (2l+1)[S_n^l - 1][S_m^l - 1]^*, \quad (17)$$

$$\sigma_i^{in} = \frac{\pi}{q^2}\sum_l (2l+1)[1 - |S_i^l|^2], \quad (18)$$

where S^l is the scattering matrix; $S_{ii}^l \equiv S_i^l$.

The final form of the system (12)—(14), with the collisions taken into account, is

$$\left[i\left(\omega - \omega_0 \pm \frac{\hbar}{m_a}k_x p\right) + \left(\frac{\Gamma}{2} + i\Delta + \nu_{mn}\right)\right]\rho_{mn}(\mathbf{k}, \mathbf{k}\pm\mathbf{p}) - \int A_{mn}(\mathbf{k}, \mathbf{k}')\rho_{mn}(\mathbf{k}', \mathbf{k}'\pm\mathbf{p})\,d\mathbf{k}' = \frac{\lambda}{2}[zW(\mathbf{k}) + \rho_n(\mathbf{k}) - \rho_m(\mathbf{k})], \quad (19)$$

$$(\beta_n + \nu_n)\rho_n(\mathbf{k}) - \int A_{nn}(\mathbf{k}, \mathbf{k}')\rho_n(\mathbf{k}')\,d\mathbf{k}' = -\operatorname{Re}\lambda^*[\rho_{mn}(\mathbf{k}, \mathbf{k}+\mathbf{p}) + \rho_{mn}(\mathbf{k}, \mathbf{k}-\mathbf{p})], \quad (20)$$

$$(\beta_m + \nu_m)\rho_m(\mathbf{k}) - \int A_{mm}(\mathbf{k}, \mathbf{k}')\rho_m(\mathbf{k}')\,d\mathbf{k}' = \operatorname{Re}\lambda^*[\rho_{mn}(\mathbf{k}, \mathbf{k}+\mathbf{p}) + \rho_{mn}(\mathbf{k}, \mathbf{k}-\mathbf{p})]. \quad (21)$$

The solution of this system in the approximation linear in the field, i.e., the contour of the usual linear absorption of the atom $I(\omega)$ was investigated in detail in[6]. We are interested only in effects that are nonlinear in the field, i.e., in a frequency region that is narrow in comparison with $\Delta\omega_D$, where the function $C(\omega)$ differs from zero (see (11)). It is easy to verify that in this frequency region the linear-absorption contour represents the usual Doppler distribution:

$$I(\omega) = \frac{m_a}{\hbar p}W\left(\frac{m_a}{\hbar p}(\omega - \omega_0)\right).$$

4. SMALL-ANGLE SCATTERING

We assume first that the collisions are characterized by relatively small scattering angles. Then, as seen from (15), $A_{ij}(\mathbf{k}, \mathbf{k}')$ has a sharp maximum at $\mathbf{k} = \mathbf{k}'$.

We represent the equilibrium velocity distribution function in the form $W(\mathbf{k}) = W_x(k_x)W_\perp(\mathbf{k}_\perp)$, where $W_x(k_x)$ is the distribution function of the projections of the wave vectors on the x direction, i.e., on the field direction, and $W_\perp(\mathbf{k}_\perp)$ is the distribution function of the projections of the wave vectors on a plane perpendicular to the x axis. We seek the solution of (19)—(21) in the form

$$\rho(\mathbf{k}) = \tilde{\rho}(k_x)W_\perp(\mathbf{k}_\perp). \quad (22)$$

When (22) is substituted in (19)—(21), the function $W_\perp(\mathbf{k}_\perp)$ can be taken outside the integral signs at the point of the maximum of the kernel $A_{ij}(\mathbf{k}, \mathbf{k}')$ at $\mathbf{k} = \mathbf{k}'$:

$$\int A_{ij}(\mathbf{k}, \mathbf{k}')W_\perp(\mathbf{k}_\perp')\tilde{\rho}(k_x')\,dk_x'\,d\mathbf{k}_\perp = W(\mathbf{k}_\perp)\int A_{ij}(\mathbf{k}_\perp, k_x, k_x')\tilde{\rho}(k_x')\,dk_x',$$

$$A_{ij}(\mathbf{k}_\perp, k_x, k_x') = \int A_{ij}(\mathbf{k}_\perp, \mathbf{k}_\perp', k_x, k_x')\,d\mathbf{k}_\perp'.$$

As already mentioned earlier, we are interested in the structure of narrow nonlinear resonances, i.e., the frequency region $|\omega - \omega_0| \ll \Delta\omega_D$. In this frequency region, the main contribution to the interaction with the field is made by atoms whose velocities are almost perpendicular to the resonator axis, $k_x \ll k_\perp$. Under this condition, it can be shown that the function $A_{ij}(\mathbf{k}_\perp, k_x, k_x')$ actually depends only on the difference $k_x - k_x' = \xi$:

$$A_{ij}(\mathbf{k}_\perp, k_x, k_x') \approx A_{ij}(\mathbf{k}_\perp, k_x - k_x') \equiv A_{ij}(\mathbf{k}_\perp, \xi).$$

Taking this circumstance into account, a solution of (19)—(21) in third-order perturbation theory in the field can be obtained by the Fourier-transformation method. We present the final results for the function $C(\omega)$ from (10), assuming for simplicity that $A_{ij}(\mathbf{k}_\perp, \xi)$ depends little on $k_\perp$, so that this dependence can be neglected, namely, $A_{ij}(\mathbf{k}_\perp, \xi) \approx A_{ij}(\xi)$. We have

$$C(\omega) = \frac{1}{2}\frac{m_a}{\hbar p}\operatorname{Re}\int_{-\infty}^{\infty}\left[\frac{1}{\beta_n + \nu_n}(1 + G_n(q)) + \frac{1}{\beta_m + \nu_m}(1 + G_m(q))\right] \times\exp\{-2(\Omega' - \varphi'(q))|q|\}[\exp\{-2i(\Omega'' - \varphi''(q))|q|\} + 1], \quad (23)$$

$$\Omega' + i\Omega'' = \frac{m_a}{\hbar p}\left[\frac{\Gamma}{2} + \operatorname{Re}\nu_{mn} + i(\omega - \omega_0 + \Delta + \operatorname{Im}\nu_{mn})\right], \quad (24)$$

$$\varphi(q) = \varphi'(q) + i\varphi''(q) = \frac{m_a}{\hbar p}\int_{-\infty}^{\infty} A_{mn}(\xi)\frac{\sin q\xi}{q\xi}\,d\xi, \quad (25)$$

$$G_i = \frac{A_{ii}(q)}{\beta_i + \nu_i - A_{ii}(q)}, \qquad A_{ii}(q) = \int A_{ii}(\xi)e^{i\xi q}d\xi. \quad (26)$$

We note that in the considered case of small-angle scattering, when $|\mathbf{k} - \mathbf{k}'| \ll k$, we have

$$\int A_{mn}(\xi)\,d\xi = \nu_{mn}, \quad \int A_{ii}(\xi)\,d\xi = \nu_i. \quad (27)$$

Let the scattering angles be limited to $\theta \ll 1$. Then at $q \ll 1/k\theta$ we obtain from (25), (26), and (27)

$$\varphi(0) = \frac{m_a}{\hbar p}\nu_{mn} \simeq \frac{\nu_{mn}k}{\Delta\omega_D}, \quad G_i(0) = \frac{\nu_i}{\beta_i}. \quad (28)$$

At $q \gg 1/k\theta$ we have

$$\varphi(q) \approx \frac{\pi}{|q|}\frac{m_a}{\hbar p}A_{mn}(k_\perp, 0) \approx \frac{\pi}{|q|}\frac{\nu_{mn}}{\Delta\omega_D\theta}, \quad G_i(q) = 0. \quad (29)$$

It is easy to see that the form of the function $C(\omega)$ depends significantly on the behavior of the exponentially damped factor in (23), and also on the form of the functions $G_i(q)$, i.e., on the ratio of the constants Γ, $\operatorname{Re}\nu_{mn}$ and β_i, ν_i. Bearing in mind the applications of greatest present interest, particularly experiments with an absorbing cell and molecular gases[3,10], let us consider the most typical case, $\beta_i \sim \nu_i$. Owing to the presence of an exponentially damped factor in the integral (23), the main contribution to this integral is made by the values $q \lesssim q_0$, where

$$q_0 = \frac{1}{\Omega' - \varphi'} \approx \frac{\hbar p}{m_a}\frac{1}{\Gamma} \approx \frac{\Delta\omega_D}{\Gamma k},$$

since (see (24)—(27))

$$\Gamma \leqslant \Omega' - \varphi'(q) \leqslant \Gamma + 2\operatorname{Re}\nu_{mn}.$$

The form of the function $C(\omega)$ depends essentially on the ratio of the quantities q_0 and $1/k\theta$.

Let us examine first the case of low densities of the perturbing particles N, when

$$q_0 \gg 1/k\theta, \quad \Gamma \ll \Delta\omega_D\theta. \quad (30)$$

To calculate $C(\omega)$ it is convenient to break up the integral (23) into two terms, $C(\omega) = C_1(\omega) + C_2(\omega)$, retaining in the first term that part of the pre-exponential

factor which does not depend on q, and retaining the function $G_i(q)$ in the second term. It follows from (30) that the function $\varphi(q)$ in the integral $C_1(\omega)$ can be set equal to (29), after which the integral is easily calculated. In the integral $C_2(\omega)$, owing to the decrease of the functions $G_i(q)$, the main contribution is always made by the values $q \lesssim 1/k\theta$. It is easily seen that under the condition (30), the argument of the decreasing exponential remains much smaller than unity in this range of q.[2)] We therefore obtain for $C(\omega)$

$$C(\omega) = C_1(\omega) + C_2(\omega), \tag{31}$$

$$C_1(\omega) = \left(\frac{1}{\beta_n + \nu_n} + \frac{1}{\beta_m + \nu_m}\right)\frac{1}{\gamma}\left\{1 + \left[\frac{\gamma^2}{4} + (\omega - \omega_0 + \delta)\gamma\,\mathrm{Im}\frac{\pi\nu_{mn}}{\Delta\omega_D\theta}\right]\left[(\omega - \omega_0 + \delta)^2 + \frac{\gamma^2}{4}\right]^{-1}\right\}, \tag{32}$$

$$C_2(\omega) = \int_0^\infty \left(\frac{G_n(q)}{\beta_n + \nu_n} + \frac{G_m(q)}{\beta_m + \nu_m}\right)\left\{1 + \cos\left[\frac{m_a}{\hbar p}(\omega - \omega_0 + \delta)q\right]\right\} dq, \tag{33}$$

$$\gamma/2 + i\delta = \Gamma/2 + \mathrm{Re}\,\nu_{mn} + i(\Delta + \mathrm{Im}\,\nu_{mn}). \tag{34}$$

As seen from (32)—(34), the spectrum consists of a narrow resonance determined by the function $C_1(\omega)$ with width $\gamma = \Gamma + 2\,\mathrm{Re}\,\nu_{mn}$ and shift $\delta = \Delta + \mathrm{Im}\,\nu_{mn}$, and a relatively broad background determined by the function $C_2(\omega)$, with a width on the order of $\hbar pk\theta/m_a \approx \Delta\omega_D\theta$. It is easy to see that the background $C_2(\omega)$ is due to the integral terms in the equations (20) and (21) for the diagonal elements of the density matrix. It should be pointed out in this connection in the general case $\sigma^{in} \sim \sigma^{el}$ it is necessary to add to (20) and (21), besides these terms, also analogous arrival terms $jk' \to mk$ and $jk' \to nk$ for arrivals from all other levels j.

We consider now another limiting case, of large densities of the perturbed particles (large Γ), when

$$q_0 \ll 1/k\theta, \quad \Gamma \gg \Delta\omega_D\theta. \tag{35}$$

When (35) is satisfied, the function $\varphi(q)$ and $G_i(q)$ take the form (28). Integrating, we again obtain formula (11) for $C(\omega)$. Thus, this case leads to the same result as the simple relaxation scheme.

5. ALMOST ISOTROPIC SCATTERING

We consider now the case of almost isotropic scattering of an atom by perturbing particles, when as a result of collision the projection of the atom velocity on the x axis change by an amount of the order of the velocity itself, $\Delta k_x \sim k_\perp$.[3)]. We substitute $\rho(\mathbf{k})$ in (19)—(21) in the form $\tilde{\rho}(k_x)W(\mathbf{k}_\perp)$. The integral term in (19)—(21) then takes the form

$$\int A_{ij}(\mathbf{k}, \mathbf{k}')\,\rho_{ij}(k')\,dk' = \int A_{ij}(\mathbf{k}_\perp, k_x, \mathbf{k}_\perp', k_x')\,W(\mathbf{k}_\perp')\,\tilde{\rho}_{ij}(k_x')\,d\mathbf{k}_\perp' dk_x'. \tag{36}$$

At $\Delta k_x \sim k_\perp$, the function $A_{ij}(\mathbf{k}_\perp, k_x, \mathbf{k}_\perp', k_x')$ is a smooth function of the arguments k_x and k_x', whereas $\tilde{\rho}(k_x)$ has a resonance at $k_x \approx |\omega - \omega_0|m_a/\hbar p \ll k_\perp$, with width $\Delta k_x \sim \Gamma m_a/\hbar p$. We can therefore represent (36) in the form

$$\int A_{ij}(\mathbf{k}, \mathbf{k}')\,\rho_{ij}(k')\,dk' = \int A_{ij}(\mathbf{k}_\perp, 0, \mathbf{k}_\perp', 0)\,W(\mathbf{k}_\perp')\,d\mathbf{k}_\perp' \times \int \tilde{\rho}_{ij}(k_x')\,dk_x' \approx \frac{\mu_{ij}}{k}\int \tilde{\rho}_{ij}(k_x')\,dk_x', \tag{37}$$

$$\mu_{ij} = \int A_{ij}(\mathbf{k}_\perp, k_x, \mathbf{k}_\perp', k_x')\,d\mathbf{k}_\perp' dk_x'. \tag{38}$$

In the case of scattering by immobile perturbing particles we have $\mu_{ij} = \nu_{ij}$. In the general case we have $\mu_{ij} \approx \nu_{ij}$ and $\nu_{ij} - \mu_{ij} \approx (m_a/m_p)\nu_{ij}$. Taking (37) into account, the solution of the system (19)—(21) leads to the following expressions for the terms of $C(\omega)$:

$$C_1(\omega) = \left(\frac{1}{\beta_n} + \frac{1}{\beta_m}\right)\frac{1}{\gamma}\left\{1 + \left[\frac{\gamma^2}{4} + (\omega - \omega_0 + \delta)\gamma\,\mathrm{Im}\frac{\pi\mu_{mn}}{\Delta\omega_D}\right]\left[(\omega - \omega_0 + \delta)^2 + \frac{\gamma^2}{4}\right]^{-1}\right\}, \tag{39}$$

$$C_2 = \frac{2\pi}{\Delta\omega_D}\left(\frac{\mu_{nn}}{\beta_n(\beta_n - \mu_{nn})} + \frac{\mu_{mm}}{\beta_m(\beta_m - \mu_{mm})}\right) \tag{40}$$

$$\gamma/2 = \Gamma/2 + \mathrm{Re}\,\nu_{mn}, \quad \delta = \Delta + \mathrm{Im}\,\nu_{mn}. \tag{41}$$

As seen from (39)—(40), the spectrum is determined in this case only by the function $C_1(\omega)$, which is completely analogous to the function (32). The function C_2 does not depend in general on the frequency and determines the constant-background level. We note that expressions (39)—(40) for $C(\omega)$ remains valid also in a wide range of pressures, so long as $\gamma \ll \Delta\omega_D$.

6. DISCUSSION OF RESULTS

The calculations given in Secs. 4 and 5 for the shape of the resonance $C(\omega)$ have led to results that differ significantly from those following form the relaxation scheme.

The shape of the resonance $C(\omega)$ is now determined by four parameters, Γ, Δ, $\mathrm{Re}\,\nu_{mn}$, and $\mathrm{Im}\,\nu_{mn}$, as against the two parameters Γ and Δ; in addition, the shape depends strongly on the concrete form of the scattering amplitudes $f_n(\theta)$ and $f_m(\theta)$. If the scattering in the collisions is mainly through small angles $\theta \ll 1$, the ratio of the parameters Γ and $\Delta\omega_D\theta$ plays an important role. At low pressures, when $\Gamma \ll \Delta\omega_D\theta$, the function $C(\omega)$ takes the form of a narrow resonance $C_1(\omega)$ (32) against the background of a broader distribution $C_2(\omega)$ (33), with a width $\Delta\omega_D\theta$. The width γ and the shift δ of the narrow resonance $C_1(\omega)$ are equal respectively to $\Gamma/2 + \mathrm{Re}\,\nu_{mn}$ and $\Delta + \mathrm{Im}\,\nu_{mn}$. In the frequency region where $C_1(\omega)$ differs from zero, the contribution of the distribution $C_2(\omega)$ is small: $C_2/C_1 \approx \Gamma/\Delta\omega_D\theta \ll 1$. We note that $C_1(\omega)$ contains an antisymmetrical increment proportional to the parameter $(\mathrm{Im}\,\nu_{mn})/\Delta\omega_D\theta$. When the pressure is increased, a complete restructuring of the contour $C(\omega)$ takes place. At $\Gamma \gg \Delta\omega_D\theta$, the background vanishes and the narrow resonance assumes the usual form of a Lorentz contour with width Γ and shift Δ. In the intermediate region $\Gamma \sim \Delta\omega_D\theta$, the resonance is described essentially by an asymmetrical function of the frequency, and its width and shift have a nonlinear dependence on the density.

In the case of almost isotropic scattering, the width and shift of the narrow resonance are linear functions of N in the entire range of pressures $\Gamma \ll \Delta\omega_D$, and the intensity of the background is practically independent of the frequency.

The main qualitative features of the nonlinear dependence of γ and δ on the pressure, which were discussed above, have a simple plysical meaning, as can be seen from the following simple considerations. At sufficiently low pressures, the atoms interacting with the field lie in the interval $|\Delta k_x| \sim \bar{k}\Gamma/\Delta\omega_D \ll \bar{k}$, where $\bar{k}$ is the average absolute value of the wave vector. If the change of k_x as a result of the scattering, $\delta k_x \sim \bar{k}\theta$, is much larger than this interval, $\Gamma \ll \Delta\omega_D\theta$, then the function $A(\mathbf{k}, \mathbf{k}')$ can be taken from outside the integral sign in the right-hand side of (19)—(21), after which it is easy to obtain

$$\gamma/2=\Gamma/2+\operatorname{Re}\nu_{mn},\quad \delta=\Delta+\operatorname{Im}\nu_{mn}. \tag{42}$$

When the pressure is increased and $k\Gamma/\Delta\omega_D$ becomes larger than $\bar{k}\theta$ ($\Gamma \gg \Delta\omega_D\theta$), the elastic collisions do not take the atoms out of the region of interaction with the field at all. We can now take outside the integral sign the relatively smooth function ρ at the point $k = k'$, after which this integral at $\theta \ll 1$ becomes equal to $\bar{\rho}(k_x)\nu_{mn}$, and the width and shift of the resonance are respectively equal to $\gamma = \Gamma$ and $\delta = \Delta$. Since the nonlinear resonances considered by us are of interest when $\Gamma \ll \Delta\omega_D$, there exists in the case $\theta \ll 1$ two pressure regions, $\Gamma \ll \Delta\omega_D\theta$ and $\Gamma \gg \Delta\omega_D$, for which formulas (32), (33) and (11) are respectively valid. If the condition $\theta \ll 1$ is not satisfied, then we can expect the values of γ and δ to be close to those in (41) for the entire pressure range $\Gamma \ll \Delta\omega_D$.

If the gas contains perturbing particles of two different types, then we get $A = A_1 + A_2$ in (19)–(21). At sufficiently low pressures $\Gamma \ll \Delta\omega_D\theta_1$, $\Delta\omega_D\theta_2$ we then have

$$\gamma/2=\Gamma/2+\operatorname{Re}(\nu_{mn}^{(1)}+\nu_{mn}^{(2)}),\quad \delta=\Delta+\operatorname{Im}(\nu_{mn}^{(1)}+\nu_{mn}^{(2)}).$$

If $\theta_1 \ll 1$ and $\theta_2 \ll 1$, then increasing the pressure again makes the formulas

$$\gamma=\Gamma,\quad \delta=\Delta$$

valid. On the other hand, if $\theta_1 \ll 1$ but $\theta_2 \sim 1$, then we obtain for $\Gamma \gg \Delta\omega_D\theta_1$

$$\gamma/2=\Gamma/2+\operatorname{Re}\nu_{mn}^{(2)},\quad \delta=\Delta+\operatorname{Im}\nu_{mn}^{(2)}.$$

Among the above-listed qualitative features of the function $C(\omega)$, greatest interest in physical applications, particularly for gas-laser frequency stabilization, is attached to the nonlinear pressure dependence of the width and especially the position of the maxima of the resonances, a dependence that occurs in the case of small-angle scattering. We therefore consider in greater detail the forms of the functions $\gamma(N)$ and $\delta(N)$ for this case. For experiments on gas-laser frequency stabilization, of greatest interest are atomic and molecular transitions for which the scattering amplitudes f_n and f_m are practically equal. It is precisely such transitions, in view of the small value of the shift Δ, which are least sensitive to density fluctuations. Putting $S_n^l \approx S_m^l$ in (16) and (17), we obtain for this case $\sigma' \gg \sigma''$ and

$$\sigma'=\frac{\pi}{q^2}\sum_l(2l+1)[1-|S_n^l|^2]\approx\sigma^{\text{in}},$$

$$\operatorname{Re}\sigma_{mn}=\frac{\pi}{q^2}\sum_l(2l+1)|1-S_n^l|^2\approx\sigma^{\text{el}}.$$

Thus, the dependence of the width of the resonance on the pressure takes the form shown in Fig. 1, and the ratio of the slopes on sections I and II is equal to[4)]

$$\frac{\Gamma+2\operatorname{Re}\nu_{mn}}{\Gamma}=\frac{\sigma^{\text{in}}+\sigma^{\text{el}}}{\sigma^{\text{el}}}. \tag{43}$$

To explain the dependence of the resonance shift on the pressure, it is necessary to specify some concrete form of the interaction. Bearing in mind neutral gases and small-angle scattering, we assume a potential that decreases in power-law fashion, $U(R) \propto R^{-k}$. Changing over in the formulas for σ' and σ_{mn} from summation over the partial waves l to integration over the impact parameters, we can estimate the corresponding integrals (see, for example,[11]). As a result we obtain for the limiting case $\sigma' \gg \sigma''$

$$\frac{\Delta}{\Delta+\operatorname{Im}\nu_{mn}}\approx\frac{1}{2}\Gamma\left(\frac{k-3}{2(k-1)}\right)\left(\frac{\sigma^{\gamma}}{\sigma^{\text{in}}}\right)^{(k-3)/2}, \tag{44}$$

where $\Gamma(x)$ is the gamma function; $\Gamma = 3$ at $k = 6$.

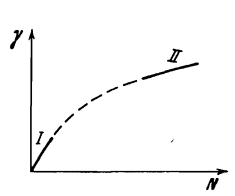

FIG. 1

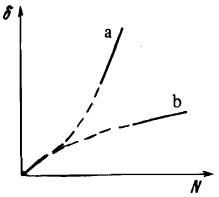

FIG. 2

FIG. 1. Dependence of the resonance width γ on the perturbing-particle density N.

FIG. 2. Dependence of the shift δ of the resonance maximum on the density of the perturbing particles N: curve a–$\sigma^{\text{in}}/\sigma^{\text{el}} < 1$ curve b–$\sigma^{\text{in}}/\sigma^{\text{el}} > 1$.

Depending on the ratio of the cross sections σ^{in} and σ^{el}, the slope of the $\delta(N)$ curve can either increase with increasing pressure ($\sigma^{\text{in}}/\sigma^{\text{el}} < 1$), or decrease ($\sigma^{\text{in}}/\sigma^{\text{el}} > 1$)—see Fig. 2. If the inelastic-scattering cross section σ^{in} is so small that the inequality $\sigma^{\text{in}} \ll |\sigma_n^{\text{el}} - \sigma_m^{\text{el}}|$ holds in spite of the condition $f_n \approx f_m$, then we can obtain from (44), putting $\sigma_m < \sigma_n$,

$$\frac{\Delta}{\Delta+\operatorname{Im}\nu_{mn}}\approx\frac{k-1}{2}\left[1-\left(\frac{\sigma_m^{\gamma}}{\sigma_n^{\gamma}}\right)^{(k-1)/2}\right]^{-(k-3)/(k-1)}.$$

Since the greatest interest attaches at present to stabilization of the $\lambda = 3.39\ \mu$ helium-neon laser frequency with an absorbing methane cell[3,10], let us compare the foregoing results with the available experimental data on methane. It was shown in[12], that for the working transition in methane the ratio of the width of the spectral line Γ to the shift Δ is anomalously large: $\Gamma/\Delta \gtrsim 100$. This situation, as noted above, can obtain in the case $f_n \approx f_m$ (see formulas (16)–(18)). Furthermore, from[3] where a nonlinear $\gamma(N)$ dependence was observed, it follows that in accordance with (43) $\sigma^{\text{in}}/\sigma^{\text{el}} \approx 1/3$ for the considered methane levels. This allows us to expect the resonance shift δ to behave with increasing N in the manner shown in Fig. 2a. Assuming $k = 6$ in (44) (van der Waals interaction), we find that the slope of the $\delta(N)$ curve should increase approximately sevenfold. There are no experimental data on the resonance shift δ in pure methane. A very recent paper[4], however, reports measurement of the dependence of δ on N in the case when methane is perturbed by xenon and helium. In both cases, the slope of $\delta(N)$ increases with pressure, just as in Fig. 2a. The larger change of the slope in the case of xenon can be attributed to the fact that the ratio $\sigma^{\text{in}}/\sigma^{\text{el}}$ is more readily smaller for xenon (heavier particle) than for helium.

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1)Equations of this type were considered also in [2,7,8].

2)From assumed relation $\beta_i \sim \nu_i$ it follows that $\operatorname{Re}\nu_{mn} \sim \Gamma$ (see (16)–(18)).

3)We recall that in the case $\gamma/\Delta\omega_D \ll 1$ considered by us the only atoms contribution to the resonance are those with $k_x \ll k_\perp$. Therefore $k = \sqrt{k_\perp^2 + k_x^2} \approx k_\perp$.

4)A similar nonlinear dependence of γ on N was experimentally observed and qualitatively explained in [3].

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