Influence of elastic collisions on nonlinear interference effects

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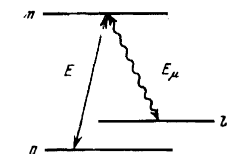
The possibility of the appearance of the Dicke effect in nonlinear ultra-high resolution optical spectroscopy is studied. The interaction between a three-level system, one of whose transitions is in the microwave region, and two optical fields is considered. The effect of elastic collisions, in which the atomic oscillator is weakly perturbed, on a resonant nonlinear interaction between the optical fields is analyzed within the framework of the strong collision model. A solution of the system of equations for the density matrix which includes the collision integral is obtained by successive approximations. It is shown that when the ground level is involved in the microwave transition, elastic collisions may lead to considerable narrowing of the nonlinear resonance within the contour of an optical spectral line. In this case the sharp structure reflects the relaxation characteristics of the microwave transition. The possibility is considered of utilizing the considered effects for nonlinear spectroscopy of ultra-high resolution, permitting one to study concurrently the characteristics of both optical and microwave transitions by optical methods.

1. INTRODUCTION

A strong electromagnetic field that is in resonance with an atomic transition alters the shape of the spectral line representing the emission and absorption of a weak field in the same transition or in an adjacent transition. Specifically, the Doppler contour of the spectral line exhibits sharp structure. The appearance of these nonlinear resonances and the dependence of their characteristics on the propagation direction of the weak field relative to the strong field (the angular anisotropy of the spectra) has been interpreted as the manifestation of nonlinear interference effects (NLIE).[1–3] The investigation of NLIE is an important branch of nonlinear spectroscopy, and has been the basis on which precision methods of nonlinear laser spectroscopy have been developed.[4,5] The resolving power of these methods is enhanced, in principle, as sharper nonlinear resonances are achieved.

There is a strong distinction between the NLIE associated with inhomogeneous and homogeneous broadening, respectively, of interacting transitions.[6] In low-pressure gases, where the spectral lines of individual atoms are broadened only through radiative relaxation, the mean free path of the atoms considerably exceeds the wavelength of the optical radiation. Consequently, the absorption and emission line of the gas as a whole undergoes considerable inhomogeneous broadening due to Doppler broadening. However, it is possible to produce conditions where without a change of velocity the atomic mean free path is considerably shorter than the wavelength of the microwave radiation. This can result from elastic collisions with the walls of the vessel or with the particles of a buffer gas without quenching an atom or essentially perturbing its phase and, therefore, without line broadening.[7] It is thus possible to exclude Doppler broadening and to reduce considerably the line width of the microwave transition (the Dicke effect [8]). In the interaction of two fields that are resonant with adjacent transitions, NLIE result from oscillations arising in the medium at a difference frequency that is close to the frequency of a forbidden transition.[3] If the wavelength of the forbidden transition is sufficiently large, then despite the absence of a field corresponding to this transition we can expect the Dicke effect to be manifested in an altered shape of the nonlinear resonances. But the line shapes of unperturbed optical transitions should not then be changed, because the wavelengths of these transitions are considerably smaller than the mean free path.

Level system of an atom interacting with the fields. The field **E** interacts with the electric dipole transition mn; the field $\mathbf{E}_\mu$ interacts with the electric dipole transition m*l*.



Collisions with a buffer gas can be accompanied by several other effects, depending on the specific atoms and transitions. It was the aim of the present work to show, within the framework of the simplest collision model encompassing the main features of the phenomena, that the Dicke effect can be manifested in ultra-high resolution nonlinear optical spectroscopy. Elastic collisions can here lead to considerable narrowing of the nonlinear resonances.

2. FUNDAMENTAL EXPRESSIONS

Let us consider the interaction of two monochromatic fields with an atom possessing the energy level system shown in the accompanying figure. The electromagnetic fields and the motion of the atomic center of inertia will be described classically. The one strong field has the amplitude **E** and frequency ω, which is close to that of the mn transition. The other, weak, field $\mathbf{E}_\mu$ has the frequency ω_μ, which is close to that of the ml transition. The corresponding matrix elements of the perturbed Hamiltonian in the interaction representation are

$$V_{mn}=Ge^{i(\Omega t-\mathbf{kr})},\quad V_{ml}=G_\mu e^{i(\Omega_\mu t-\mathbf{k}_\mu\mathbf{r})}. \tag{2.1}$$

Here $G=-d_{nm}E/2\hbar$, $G_\mu=-d_{lm}E_\mu/2\hbar$; $\Omega=\omega-\omega_{mn}$, $\Omega_\mu=\omega_\mu-\omega_{ml}$; d_{ij} is the matrix element of the dipole moment of a transition. To analyze the line shape for absorption of the weak field $\mathbf{E}_\mu$ in the presence of the strong field **E** we shall begin with the formula for the field-$\mathbf{E}_\mu$-absorption power per unit volume:

$$W_{ml}(\Omega_\mu)=\hbar\omega_\mu\langle\overline{2\operatorname{Re}\{iV_{lm}\rho_{ml}\}}\rangle_v. \tag{2.2}$$

Here ρ_{ml} is an off-diagonal element of the density matrix in the interaction representation, normalized to unit volume and averaged over an ensemble.[9,10] The bar denotes time and volume averaging; the angular brackets denote averaging over the velocities of atoms interacting with the field.

The equation for the density matrix ρ in the interaction representation is

$$\hat{L}\hat{\rho}=-i[\hat{V}\hat{\rho}]+R(\hat{\rho})+(d\rho/dt)^{\text{col}}. \tag{2.3}$$

Here $\hat{L}=d/dt+\mathbf{v}\nabla$, where $\mathbf{v}$ is the velocity of the atomic center of inertia, and $R(\hat{\rho})$ describes the radiative relaxation:

$$\begin{gathered}R_{mm}=-\Gamma_{mm}\rho_{mm},\quad R_{ll}=-\Gamma_l\rho_{ll}+\gamma_{ml}\rho_{mm},\quad \Gamma_l=\gamma_{ln},\\ R_{ij}=-\Gamma_{ij}\rho_{ij}\quad (\Gamma_{ij}=\Gamma_{ji}=(\Gamma_i+\Gamma_j)/2;\quad i,\ j=n,\ l,\ m).\end{gathered} \tag{2.4}$$

Here $\Gamma_m=\gamma_{mn}+\gamma_{ml}$, with γ_{ij} representing the probability of a transition per unit time from level i to level j; Γ_{ij} is the radiative relaxation constant of an off-diagonal element of the density matrix (the radiation linewidth). Taking the level n to be the ground level, the solution for ρ_{nn} is obtained from the conservation law

$$\operatorname{Sp}\rho(\mathbf{v})=\sum_i\rho_{ii}(\mathbf{v})=NW(\mathbf{v}),$$

where N is the total concentration of atoms having the given level structure, and $W(\mathbf{v})=(\sqrt{\pi}\,\bar{v})^{-3}\exp[-v^2/\bar{v}^2]$ is the Maxwellian velocity distribution.

The term $(d\rho/dt)^{\text{col}}$ in (2.3) takes account of elastic and inelastic collisions with particles of the buffer gas. In the general case this term can be represented by [11-14]

$$\begin{gathered}\left(\frac{d\rho}{dt}\right)^{\text{col}}_{ij(i\neq j)}=-(\Gamma_{ij}^{\text{col}}+i\Delta_{ij}+\nu_{ij})\rho_{ij}(\mathbf{v})+\int d\mathbf{v}'A_{ij}(\mathbf{v},\mathbf{v}')\rho_{ij}(\mathbf{v}'),\\ \left(\frac{d\rho}{dt}\right)^{\text{col}}_{mm}=-(\beta_m+\nu_m)\rho_{mm}(\mathbf{v})+\int d\mathbf{v}'A_{mm}(\mathbf{v},\mathbf{v}')\rho_{mm}(\mathbf{v}').\end{gathered} \tag{2.5}$$

Since the equilibrium population of level l is not zero and corresponds to the Boltzmann distribution, in the equation for ρ_{ll} we also take into account the possibility of transitions from level n to level l, induced by collisions:

$$\left(\frac{d\rho}{dt}\right)^{\text{col}}_{ll}=-(\beta_l+\nu_l)\rho_{ll}(\mathbf{v})+\int d\mathbf{v}'A_{ll}(\mathbf{v},\mathbf{v}')\rho_{ll}(\mathbf{v}')+\int d\mathbf{v}'A_n^l(\mathbf{v},\mathbf{v}')\rho_{nn}(\mathbf{v}'). \tag{2.6}$$

Here Γ_{ij}^{col} and Δ_{ij} are the collisional broadening and shift; β_i is the frequency of inelastic, and ν_i is the frequency of elastic collisions in the state i; ν_{ij} plays the role of the frequency of elastic collisions for off-diagonal elements. The kernels of the collision integrals, $A(\mathbf{v},\mathbf{v}')$, signify the probabilities of a velocity change $\mathbf{v}'\to\mathbf{v}$ per unit time. The indicated parameters can be selected in accordance with the model;[11-13] in [14] they are expressed in terms of the characteristics of elementary elastic and inelastic scattering events. Specifically, ν_i and A_{ii} are defined in terms of the squared modulus of the scattering amplitude, $|f_i|^2$, while ν_{ij} and A_{ij} are defined in terms of the $f_if_j^*$ products. Thus in the general case the frequencies of collisions and the kernels are complex; this reflects the shift of atomic resonances that is induced by elastic collisions.

We shall now consider the simplest analyzable case, that of almost isotropic scattering, which yields the effects of interest. In such collisions the projection of the atomic velocity on any direction is changed by the same order of magnitude as the velocity. For subsequent calculations we shall specify the forms of the kernels A_{ii}, A_{ij}, and A_n^l, using the strong collision model.[11-13] This model is based on the assumption that $A(\mathbf{v},\mathbf{v}')$ is independent of $\mathbf{v}'$, i.e.,

$$A(\mathbf{v},\mathbf{v}')=A(\mathbf{v}). \tag{2.7}$$

In other words, it is assumed that the velocity $\mathbf{v}$ of a particle after a collision does not depend on its velocity $\mathbf{v}'$ before the collision. This model reflects the fundamental qualitative characteristics of the scattering of light particles by heavy particles.

A strong electromagnetic field that interacts resonantly with gas atoms induces transitions between their energy levels. Since nonlinear resonances occur when $\Omega-\mathbf{kv}=0$, in isotropic scattering the strongest deviation from the equilibrium distribution results only from the projection of atomic velocities on the strong-field wave-vector direction, such as v_z. These velocities, which lie within a narrow range, are responsible for nonlinear resonances in the weak-field absorption power.

We represent $\rho(\mathbf{v})$ as $\rho(v_z)W(v_\perp)$. Integrating the equations for $\rho(\mathbf{v})$ over the velocity projections on directions orthogonal to $\mathbf{k}$ and neglecting the dependence of Γ^{col}, Δ, ν, and β on v_z in the nonlinear-resonance region, we obtain an approximate system of equations for $\rho(v_z)$. These equations have the form of (2.3), where the term $\mathbf{v}\nabla$ is rellaced by $v_z d/dz$, while Γ^{col}, Δ, ν, and β in (2.5) and (2.6) are replaced approximately by averages over the atomic velocities. The assumption (2.7) and the requirement $(d\rho/dt)^{\text{col}}=0$ under equilibrium conditions in the absence of inelastic collisions lead to

$$\begin{gathered}\int A_{ij}(\mathbf{v},\mathbf{v}')\,d\mathbf{v}_\perp'=\bar{A}_{ij}(v_z)=\nu_{ij}W(v_z),\\ \bar{A}_{ii}(v_z)=\nu_iW(v_z),\quad A_n^l(v_z)=\nu_n^lW(v_z),\end{gathered} \tag{2.8}$$

where $W(v_z)$ is the Maxwellian distribution of atomic-velocity projections on the wave-vector ($\mathbf{k}$) direction of the strong field. The derived system of equations enables us to consider the Dicke effect qualitatively in nonlinear optical spectroscopy.

Since the absence of fields should be accompanied by an equilibrium distribution of the populations, from (2.3) in conjunction with (2.4)–(2.6) and (2.10) we obtain

$$N_l/N_n=\nu_n^l/\gamma_{ln}=\exp[-\hbar\omega_{ln}/K_BT], \tag{2.9}$$

where N_l and N_n are the equilibrium population levels and K_B is the Boltzmann constant.

The system of equations which is derived from (2.3) for the density matrix is solved in the stationary case by successive approximations to the first order in G_μ and to the second order in G. In this approximation, with the aid of (2.2), for the case of parallel $\mathbf{k}$ and $\mathbf{k}_\mu$ we obtain

$$\begin{aligned}W_{ml}(\bar{\Omega}_\mu)=2\hbar\omega_\mu N_l|G_\mu|^2\operatorname{Re}\Big\{&\eta_{ml}z_{ml}+\frac{1}{\bar{\Gamma}_l}\Big[az_{ml}\operatorname{Re}\eta_{mn}z_{mn}\\ &+b\left(\eta_{mn}\frac{(k_\mu/k)z_{ml}-z_{mn}}{(k_\mu/k)p_{mn}-p_{ml}}+\eta_{mn}^*\frac{(k_\mu/k)z_{ml}+z_{mn}^*}{(k_\mu/k)p_{mn}^*+p_{ml}}\right)\Big]\frac{N_n}{N_l}|G|^2\\ &-\eta_{mn}^*\frac{(k_\mu/\Delta k)z_{ml}+z_{ln}^*}{(p_{mn}^*-(k/\Delta k)p_{ln}^*)(p_{ml}+(k_\mu/\Delta k)p_{ln}^*)}\left[1+\eta_{ln}^*\nu_{ln}^*\left(z_{ln}^*-\frac{k}{\Delta k}z_{mn}^*\right)\right]\\ &\times\frac{N_n}{N_l}|G|^2+\eta_{mn}^*\frac{(k_\mu/k)z_{ml}+z_{mn}^*}{((\Delta k/k)p_{mn}^*-p_{ln}^*)((k_\mu/k)p_{mn}^*+p_{ml})}\\ &\times\frac{N_n}{N_l}|G|^2-\eta_{ml}^2\frac{(k_\mu/\Delta k)z_{ml}+z_{ln}^*}{(p_{ml}+(k_\mu/\Delta k)p_{ln}^*)^2}\Big[1-\frac{p_{ml}+(k_\mu/\Delta k)p_{ln}^*}{z_{ml}+(\Delta k/k_\mu)z_{ln}^*}\frac{\partial z_{ml}}{\partial p_{ml}}\\ &+\eta_{ln}^*\nu_{ln}^*\left(z_{ln}^*+\frac{k_\mu}{\Delta k}z_{ml}\right)\Big]|G|^2\Big\}.\end{aligned} \tag{2.10}$$

Here the following notation has been used:

$$\begin{gathered}\Delta k=k-k_\mu,\quad \eta_{ij}=(1-\nu_{ij}z_{ij})^{-1},\quad p_{mm}=\bar{\Gamma}_{mn}-i\bar{\Omega},\quad p_{ln}=\bar{\Gamma}_{ln}-i(\bar{\Omega}-\bar{\Omega}_\mu);\\ \bar{\Gamma}_j=\Gamma_j+\beta_j+\nu_j,\quad \tilde{\Gamma}_j=\Gamma_j+\beta_j,\quad \bar{\Omega}_{ij}=\Omega_{ij}+\Delta_{ij}+\nu_{ij}'';\end{gathered}$$

$$a=\frac{\nu_m}{\tilde{\Gamma}_m}\frac{\gamma_{ml}-\bar{\Gamma}_l}{\bar{\Gamma}_m}+\frac{\nu_n^l-\nu_l}{\tilde{\Gamma}_l+\nu_n^l}\frac{\gamma_{ml}-\nu_n^l}{\tilde{\Gamma}_m},\qquad b=\frac{\gamma_{ml}-\bar{\Gamma}_l}{\bar{\Gamma}_m};$$

$$z_{ml}=\int_{-\infty}^{\infty}\frac{dvW(v)}{p_{ml}+ik_\mu v},\quad z_{mn}=\int_{-\infty}^{\infty}\frac{dvW(v)}{p_{mn}+ikv},\quad z_{ln}=\int_{-\infty}^{\infty}\frac{dvW(v)}{p_{ln}+i\Delta kv}.$$

We can write approximately:

$$z_{ln}=\begin{cases}\dfrac{\sqrt{\pi}}{\Delta k\bar{v}}\exp\left\{-\left(\dfrac{\Omega-\Omega_\mu}{\Delta k\bar{v}}\right)^2\right\}\left[1+iI\left(\dfrac{\Omega-\Omega_\mu}{\Delta k\bar{v}}\right)\right] & \text{if } \bar{\Gamma}_{ln}\ll\Delta k\bar{v},\\ 1/p_{ln} & \text{if } \bar{\Gamma}_{ln}\gg\Delta k\bar{v}.\end{cases}$$

Here

$$I(x)=\frac{2}{\sqrt{\pi}}\int_0^x e^{t^2}dt.$$

In the case of antiparallel $\mathbf{k}$ and $\mathbf{k}_\mu$ the quantity Δk becomes equal to $k+k_\mu$, and in (2.10) we must replace k_μ everywhere by $-k_\mu$. The equation is applicable for $|G|^2\ll\bar{\Gamma}_{ln}\bar{\Gamma}_{mn}$.

The terms in the first square bracket reflect the change in absorption of the weak field resulting from the change in level populations that is induced by the field $\mathbf{E}$. The part proportional to b results from the change in level populations that is induced by the strong field because of selective interaction with atoms at specific velocities. The part proportional to a results from nonselective interaction, which means that elastic collisions also involve atoms of different velocities in the interaction with the field. The second and third groups of terms arise from the oscillations induced in the medium at the difference frequency $\omega-\omega_\mu$. These terms determine the NLIE and splitting. In the given perturbation approximation these terms can be interpreted like those resulting from the interference of stepwise and two-photon transitions in which $\hbar\omega_\mu$ photons participate.[2]

3. LINE SHAPE ANALYSIS

For the purpose of analyzing the line-shape change caused by elastic collisions, we first examine the line-shape expression in the absence of elastic collisions. This case corresponds to the frequently employed relaxation-constant model. This model can be used for considering the Weisskopf broadening mechanism, the Lorentz model (collisional quenching), and radiative relaxation at low pressures. The corresponding line shape can be observed in the absence of a buffer gas. The value $\nu_{ij}=0$ can also be realized for the case of a vanishing scattering amplitude in one of the states i or j. As ν_c, ν_{ij}, and ν_h^l approach zero, from the general expression (2.10) with $\bar{\Gamma}_{mn}\ll k\bar{v}$ and $\bar{\Gamma}_{ml}\ll k_\mu\bar{v}$ we obtain

$$\begin{aligned}W_{ml}(\bar{\Omega}_\mu)=W_{ml}(0)\exp\left\{-\left(\frac{\bar{\Omega}_\mu}{k_\mu\bar{v}}\right)^2\right\}\Bigg\{1+2b\frac{k_\mu}{k}\frac{\bar{\Gamma}_-}{\bar{\Gamma}_l}\\ \times\frac{|G|^2}{\bar{\Gamma}_-^2+(\bar{\Omega}_\mu\mp(k_\mu/k)\bar{\Omega})^2}\frac{N_n}{N_l}-\theta(\mathbf{k}_\mu\mathbf{k})\frac{2k_\mu}{k}\frac{1}{\bar{\Gamma}}\\ \times\left[\frac{\bar{\Gamma}_+}{\bar{\Gamma}_+^2+(\bar{\Omega}_\mu-(k_\mu/k)\bar{\Omega})^2}-\frac{\bar{\Gamma}_-}{\bar{\Gamma}_-^2+(\bar{\Omega}_\mu-(k_\mu/k)\bar{\Omega})^2}\right]|G|^2\frac{N_n}{N_l}\\ -\theta(\mathbf{k}_\mu\mathbf{k}-|\mathbf{k}_\mu|^2)\,2\frac{\Delta k}{k}\frac{k_\mu}{k}\frac{\bar{\Gamma}_+^2+(\bar{\Omega}_\mu-(k_\mu/k)\bar{\Omega})^2}{[\bar{\Gamma}_+^2+(\bar{\Omega}_\mu-(k_\mu/k)\bar{\Omega})^2]^2}|G|^2\Bigg\},\end{aligned}\tag{3.1}$$

where

$$W_{ml}(0)=2\hbar\omega_\mu\frac{\sqrt{\pi}}{k\bar{v}}N_l|G_\mu|^2,\qquad \bar{\Gamma}_-=\bar{\Gamma}_{ml}+\frac{k_\mu}{k}\bar{\Gamma}_{mn},$$

$$\bar{\Gamma}=\bar{\Gamma}_{mn}+\bar{\Gamma}_{ml}-\bar{\Gamma}_{ln},$$

$$\bar{\Gamma}_+=\begin{cases}\dfrac{k_\mu}{k}\bar{\Gamma}_{ln}+\dfrac{\Delta k}{k}\bar{\Gamma}_{ml}, & \text{if } k_\mu<k,\\ \bar{\Gamma}_{ln}+\dfrac{\Delta k}{k}\bar{\Gamma}_{mn}, & \text{if } k_\mu>k,\end{cases}$$

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

In the curly brackets of (3.1) the first term which is proportional to $|G|^2$ reflects the change in absorption that results from a change of population. The integral with respect to $\bar{\Omega}_\mu$ of the second group of terms in the rectangular brackets vanishes; these terms reflect NLIE. The last term is due to splitting.[15,16] The equation indicates the strong angular anisotropy of the spectral manifestations of nonlinear resonance processes. As an example, for Lorentz collisions with purely radiative relaxation, in parallel waves when $\bar{\Gamma}_{ij}=(\bar{\Gamma}_i+\bar{\Gamma}_j)/2$ the structures having the width $\bar{\Gamma}_-$ cancel out if $\gamma_{ml}\ll\bar{\Gamma}_l$ and there remains only a sharp structure of width $\bar{\Gamma}_+\ll\bar{\Gamma}_-$ and having its center at $\bar{\Omega}_\mu=(k_\mu/k)\bar{\Omega}$ on the background of the Doppler absorption curve. For $\Delta k\ll k$ there is negligibly small splitting. In antiparallel waves there remains only a structure of width $\bar{\Gamma}_-$ with its center at $\bar{\Omega}_\mu=-(k_\mu/k)\bar{\Omega}$. Hence it is possible to determine directly and concurrently the relaxation properties of both the optical and microwave subsystems by the methods of optical spectroscopy, when the widths $\bar{\Gamma}_-$ and $\bar{\Gamma}_+$ depend mainly on the relaxations properties of these subsystems. Since absorption in optical transitions considerably exceeds that in microwave transitions, measurements can be performed at low pressures. However, at the very low pressures $p\lesssim10^{-3}$ Torr the width $\bar{\Gamma}_+$ begins to be determined mainly by the "optical addition" $(\Delta k/k)\bar{\Gamma}_{ml}$ or $(\Delta k/k)\bar{\Gamma}_{mn}$. This fact prevents utilization of the given method at very low pressures. The situation changes, however, when a buffer gas is used.

To simplify the analysis of the general formula (2.10) in the presence of elastic collisions, let us consider the case where the scattering amplitudes in the states n and l are identical. According to [14], we can then assume that ν_{ln} is real and that the broadening and shifts in optical transitions are identical.

If $\nu_{ln}\gg\Delta k\bar{v}$ the atomic mean free path $l=\bar{v}/\nu_{ln}$ is determined mainly by the elastic collisions and is considerably smaller than the wavelength of the microwave transition. Since the interacting optical transitions are linked to the microwave transition due to the contribution of two-photon processes, the general formula reflects the dependence of the spectral manifestations of nonlinear processes on the type of broadening of the latter transition. Indeed, in the considered case when $\mathbf{k}_\mu$ and $\mathbf{k}$ are parallel we have $z_{ln}=\bar{p}_{ln}^{-1}$ and

$$\eta_{ln}z_{ln}\approx\frac{1}{\tilde{\Gamma}_{ln}+i(\bar{\Omega}_\mu-\bar{\Omega})};\qquad \tilde{\Gamma}_{ln}\ll\bar{\Gamma}_{ln},\qquad \tilde{\Gamma}_{ln}=\bar{\Gamma}_{ln}-\nu_{ln}.\tag{3.2}$$

This expression reflects the increase of the coherence time of oscillations at the resonance frequency,[11,12] due to the fact that the phases are slightly perturbed by collisions. The separate trains into which the oscillations of the atomic oscillator are divided are coherent. Their interference can be neglected only for trains of length $l\gg\lambda_{ln}/2\pi$; however, this means that $\nu_{ln}\ll\Delta k\bar{v}$. Since $|\nu_{mn}|\ll k\bar{v}$ and $|\nu_{ml}|\ll k_\mu\bar{v}$, the line shapes of the optical transitions remain practically unchanged and

$$\eta_{mn}z_{mn}\approx z_{mn},\qquad \eta_{mn}z_{mn}\approx z_{mn}.$$

We shall hereafter neglect the difference between the vectors k_μ and k. Thus for parallel waves in the considered case, with Ω_μ in the region $|\bar{\Omega}_\mu - \bar{\Omega}| \ll \nu_{ln}$ and $\hbar\omega_{ln}/k_B T \ll 1$, from (2.12) we obtain

$$W_{ml}(\bar{\Omega}_\mu) = W_{ml}(0) \exp\left\{-\left(\frac{\bar{\Omega}_\mu}{k_\mu \bar{v}}\right)^2\right\}\left\{1 + b\frac{|G|^2}{\bar{\Gamma}_l \bar{\Gamma}_-}\right.$$
$$+ \frac{2\Gamma_{ln}|G|^2}{\bar{\Gamma}[\bar{\Gamma}_{ln}^2 + (\bar{\Omega}_\mu - \bar{\Omega})^2]} + \frac{\sqrt{\pi}}{k\bar{v}} e^{-(\bar{\Omega}/k\bar{v})^2}\left[\left(\frac{\hbar\omega_{ln}}{K_B T} + 8I^2\left(\frac{\bar{\Omega}}{k\bar{v}}\right)\right)\right. \quad (3.3)$$
$$\left.\left.\times \frac{\tilde{\Gamma}_{ln}}{\tilde{\Gamma}_{ln}^2 + (\bar{\Omega}_\mu - \bar{\Omega})^2} + 2I\left(\frac{\bar{\Omega}}{k\bar{v}}\right)\frac{\bar{\Omega}_\mu - \bar{\Omega}}{\tilde{\Gamma}_{ln}^2 + (\bar{\Omega}_\mu - \bar{\Omega})^2}\right]|G|^2\right\}.$$

The spectral structure on the considered portion of the Doppler curve results from NLIE (the third term in the curvy brackets) and from the combination of NLIE and splitting (the expression in square brackets). The term that is proportional to b results from selective, with respect to velocity, change of the level populations, induced by the field **E**. The corresponding structure possesses width of the order of $\bar{\Gamma}_- \gg \bar{\Gamma}_{ln}$. When Ω_μ is changed by an amount of the order of ν_{ln} we neglect the frequency dependence of this term. The term proportional to a is omitted, because in the present case it is negligibly small as compared with the retained terms. It follows from (3.3) that elastic collisions with phase memory lead to a qualitative change in the form of the nonlinear resonance. Thus for parallel waves, in addition to the structure with the width $\bar{\Gamma}_{ln}$ there appears an extremely narrow structure with width $\tilde{\Gamma}_{ln} \ll \bar{\Gamma}_{ln}$. If $\bar{\Omega} = 0$, then $I = 0$ and this spectral structure has a simple Lorentz form. Its width for forbidden transitions is determined mainly by quenching collisions and can be much smaller than $\Delta k\bar{v}$ and $\bar{\Gamma}_{ln}$. Another remarkable property consists in the fact that in the considered case the splitting ceases to depend on the ratio of k and k_μ and can occur even for $\Delta k \ll k$.

For antiparallel waves $|k - k_\mu|$ must be replaced by $k + k_\mu \gg |k - k_\mu|$. In this case the conditions for the Dicke effect are not fulfilled, because $(k + k_\mu)\bar{v} \gg \nu_{ln}$. As previously, the line shape is that of a Doppler curve with a nonlinear addition and now consists of a "band" which depends slightly on Ω_μ and is proportional to a $\exp[-(\Omega/k\bar{v})^2]$, and the contour is of width $\bar{\Gamma}_-$ with its center at $\Omega_\mu = -\bar{\Omega}$.

It follows from (3.3) that the amplitudes of the structures with widths $\tilde{\Gamma}_{ln}$ and $\bar{\Gamma}_{ln}$ have the ratio

$$\frac{(\hbar\omega_{ln}/K_B T)}{(\bar{\Gamma}\bar{\Gamma}_{ln}/\tilde{\Gamma}_{ln} k\bar{v})}.$$

with $\lambda_{ln} = 1$ cm, $\hbar\omega_{ln}/K_B T \sim 10^{-3}$, $\bar{\Gamma}_{ln} \sim 10^2 \Delta k\bar{v}$, $\bar{\Gamma} \sim \bar{\Gamma}_m \sim 10^7$ sec^{-1}, and $\bar{\Gamma}_n \sim 10^2$ sec^{-1} [16] this ratio is of the order of unity. To observe the considered effects it is sufficient to have a field intensity at which the condition $|G^2| \sim \bar{\Gamma}\bar{\Gamma}_{ln}$ is satisfied. These values are smaller by the factor $\bar{\Gamma}/\bar{\Gamma}_{ln} \sim 10$ than the values usually employed when NLIE are investigated for excited levels. For a microwave transition between two optically excited states the utilization of a buffer gas makes no essential difference, because in this case the transition is broadened by radiative relaxation even without the presence of a buffer gas ($\Gamma_{ln} \sim \Gamma_{mn}$, Γ_{ml}).

4. CONCLUSION

On the basis of the foregoing analysis we may conclude that "image transfer" of the line shape of a microwave transition into the optical region is possible. Nonlinear optical spectroscopy can then be used to study in a uniform manner the relaxation properties of the two different atomic subsystems—the optical and the microwave. The resolving power of the method is determined by $\tilde{\Gamma}_{ln}$ and can be considerably smaller than the natural line widths of the optical transitions.

Increasing partial pressure of a buffer gas is accompanied by greater frequency of elastic collisions, and the shape of a nonlinear resonance then changes in accordance with (3.1) and (3.3).

In this work we have used a simple parametrization of the term $(d\rho/dt)^{col}$ in the equation for the density matrix, which describes qualitatively the principal properties of the scattering of light particles by heavy particles. It should be noted that in the case of small-angle scattering (scattering by light particles) the characteristics of nonlinear resonance can exhibit a more complicated dependence on the buffer gas pressure, as has been found for the pressure dependence of the width and shift of the Lamb dip.[14]

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91