Collective effects in resonant fluorescence

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Resonant scattering of intense radiation by a system of atoms in a volume with linear dimensions (small, compared to the wavelength of the scattered radiation) is considered. Expressions for the stationary density matrix, which are valid for any number N of scattering particles, are obtained for the cases of exact resonance and of widely spaced components. It is shown that the previously predicted hysteresis does not occur in the present system. At exact resonance and in the limit as $N \to \infty$ the system undergoes a sharp transition (similar to a second-order transition) from the region of classical behavior into a region in which quantum properties are significant. At short delay times τ and when the number of scattering atoms exceeds two, the intensity correlation function is a growing function of τ, i.e., the photon antibunching effect that takes place in monatomic fluorescence is now absent. At $N > 1$ the intensity correlation function contains a new component (compared with the case $N = 1$) at a frequency 2Ω (Ω is the distance between the central and lateral spectral components).

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1. The problem of resonant fluorescence (or resonant scattering of light) was among the first to be solved by using quantum-electrodynamics methods. The advent of lasers has again attracted attention to this problem. It was shown in many theoretical and experimental studies (see the literature in Ref. 1) that sideband components appear in the scattering spectrum in the field of intense radiation. Singularities were also observed in the statistics of the scattered light.[2-5] Until recently, however, all the theoretical calculations of the resonant fluorescence were made under the assumption that each atom radiates independently of its surrounding atoms. This assumption, while well satisfied in the case of very low atom densities and corresponding to a neglect of the action exerted by the radiation of one atom on that of the others, does not hold, of course, if the average distance between the atoms is much less than the radiation wavelength. The collective effects produced when the last condition is satisfied were investigated mainly as applied to spontaneous decay of excited states of atoms (see Refs. 6–9 as well as the literature cited in Ref. 9).

A number of recent studies[10-16] have dealt with the manifestation of collective effects in resonant scattering of intense radiation. The basis for the analysis of these effect is the equation of motion of the density matrix of a system of N two-level atoms contained in a volume whose linear dimensions are much less than the wavelength of the scattered light:

$$d\hat{\rho}/dt = i[\hat{V}, \hat{\rho}] + (\gamma/2)\{[J_-, \hat{\rho}J_+] + [J_-\hat{\rho}, J_+]\}.$$

Equation (1) differs from the usual equation for the description of superradiance in that it contains the term $i[\hat{V}, \hat{\rho}]$. Here

$$\hat{V} = v(J_+ + J_-) + \varepsilon J_z \tag{2}$$

is the operator of the interaction of the radiation with the atoms (in the interaction representation); $v = \mathbf{p} \cdot \mathbf{E}/\hbar$ is the matrix element of the dipole interaction between the incident radiation of amplitude E_0 and frequency ω_0 with the resonant transition; $\varepsilon = \omega_0 - \omega$ is the detuning from the resonant frequency; γ is the probability of spontaneous transitions; $\hat{J}_+, \hat{J}_-$ are the collective transi-

tion operators and satisfy the commutation relations of the angular-momentum operators:

$$[J_+, J_-] = 2J_z, \quad [J_z, J_{\pm}] = \pm J_{\pm}. \tag{3}$$

It is easily seen that Eq. (1) conserves the square of the angular momentum

$$\langle J^2 \rangle = \langle J_{\pm}J_{\mp} \rangle + \langle J_z^2 \rangle \mp \langle J_z \rangle = j(j+1), \tag{4}$$

where $2J \equiv N$ is the number of emitted atoms.

The problem (1) was solved in Refs. 10–12 for the case of several atoms ($N = 2, 3$). It is much more difficult to deal with the case of many atoms ($N \gg 1$) because of the mathematical complexity of the problem. Various solution methods were used—expansion of the density operator in terms of coherent spin states, solution of the equations for the conditional quasiprobability function,[13] solution of the corresponding stochastic equations by the averaging method,[15] numerical analysis for $N \leqslant 15$,[15] and the method of factoring the mean values.[16,17] However, although a variety of methods was resorted to, the problem of cooperative resonant fluorescence did not find its final solution, and the results obtained by various authors are contradictory in many respects. In particular, the use of the factoring method has led the authors of the cited papers[16,17] to state that hysteresis is present in resonant fluorescence at $N \geqslant 8$. On the other hand, from the equations derived for the coefficients of the expansion of the density matrix in terms of the eigenstates of the operator $\hat{J}_z$, it is easy to see that the determinant of the right-hand side does not vanish at any value of the parameter v/γ. Consequently, to each value of v/γ corresponds a unique stationary solution. In other words, there is no hysteresis in such a system and the use of the factoring method within the framework of the model described by (1) is invalid.

To examine the collective effects in resonant fluorescence we single out in this article two cases for which stationary solutions have been obtained for Eq. (1) and are valid at any number N of emitted atom. The first corresponds to exact resonance ($\varepsilon = 0$). The second corresponds to the situation wherein the spectrum consists of individual non-overlapping components. We

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prove the existence at $\varepsilon = 0$, in the asymptotic limit $N \rightarrow \infty$, of a critical value of the power of the incident radiation, up to which the system exhibits classical (coherent) properties, and above which quantum effects become significant. We analyze the spectrum and the two-time correlation function of the intensity of the scattered light. It is shown that at small time delays and at $N > 2$ the correlation of the intensities is a decreasing function of the delay, i.e., in this case the photon antibunching effect, which takes place in monatomic fluorescence, is absent. An additional component appears in the intensity fluctuation spectrum at $N > 1$.

2. In the case of zero frequency detuning ($\varepsilon = 0$) Eq. (1) has an exact stationary solution. But first we obtain an approximate solution that is valid in a sufficiently large range of variation of the scattered-light power.

If we put

$$\hat{J}_+ = J_+ - i\bar{v}, \quad \hat{J}_- = J_- + i\bar{v}, \tag{5}$$

then Eq. (1) reduces to the form

$$d\hat{\rho}/d\tau = [\hat{J}_-, \hat{\rho}\hat{J}_+] + [\hat{J}_-\hat{\rho}, \hat{J}_+], \tag{6}$$

where $\bar{v} = 2v/\gamma$, $\tau = \gamma t/2$. From (6), by virtue of its analogy with the usual equation of spontaneous decay for the spin j, it follows that if the operator $\hat{J}_-$ were to have eigenvalues, then the stationary solution of (1) would be

$$\hat{\rho}_{st} = |\tilde{J}_-\rangle\langle\tilde{J}_-|, \tag{7}$$

where the state $|\tilde{J}_-\rangle$ would be determined from the relation

$$\hat{J}_-|\tilde{J}_-\rangle = 0. \tag{8}$$

Since the spectrum of $\hat{J}_z$ is limited, the operator $\hat{J}_-$ has no exact eigenvalues. However, putting

$$|\tilde{J}_-\rangle = \sum_{n=0}^{2j} c_{-j+n}|-j+n\rangle, \tag{9}$$

where

$$c_{-j+n} = A^{-\frac{1}{2}}(-i\bar{v})^n[(2j-n)!/n!(2j)!]^{\frac{1}{2}}, \tag{10}$$

$$A = \sum_{n=0}^{2j} \bar{v}^{2n}(2j-n)!/n!(2j)!,$$

we have

$$\hat{J}_-|\tilde{J}_-\rangle = i\bar{v}c_j|j\rangle. \tag{11}$$

Thus, the state $|\tilde{J}_-\rangle$ defined by formulas (9) and (10) at

$$|\bar{v}c_j|^2 \ll 1 \tag{12}$$

is an almost exact eigenstate of the operator $\hat{J}_-$, and the density matrix (7) is an almost exact stationary solution of Eq. (1). At small j the inequality (12) is satisfied at small $\bar{v}$. At appreciable j the range of values of $\bar{v}$ at which (12) is valid becomes larger. In fact, using in this case Stirling's formula for $(2j)!$ we can easily show that

$$|\bar{v}c_j|^2 < (1/2\pi)(\bar{v}e/2j)^{4j}(\bar{v}/2j)^2. \tag{13}$$

The last expression remains much less than unity at

$$\bar{v}e/2j < 1 - 1/2j. \tag{14}$$

From this we find that the region in which the solution (7) is valid broadens with increasing j to values of $\bar{v}$ on the order of $2j/e$.

It should be noted here again that the expansion of the solution (7) in powers of $\bar{v}$ coincides with the exact expansion (which can be obtained by perturbation theory) up to terms of order $4j$.

By virtue of (11) and of the relation

$$\langle J_+ J_- \rangle = \bar{v}\langle J_v \rangle, \tag{15}$$

which follows from (1), it follows that when (12) is satisfied the following equation holds:

$$\langle J_+ J_- \rangle = \langle J_+ \rangle \langle J_- \rangle = \bar{v}^2, \tag{16}$$

i.e., the solution (7) is quasiclassical. Recognizing that the integrated intensity of the scattered light is proportional to $\langle \hat{J}_+\hat{J}_- \rangle$, and the intensity of the coherent δ-like component is proportional to $\langle \hat{J}_+ \rangle\langle \hat{J}_- \rangle$ (see, e.g., Ref. 1), we find from (16) that in this approximation the entire scattered radiation is coherent. By virtue of the remark made above, the noncoherent component appears in the scattered radiation only in the $(2j+1)$st order in the intensity of the exciting radiation.

Thus, with increasing j the "region of coherence" with respect to the parameter $\bar{v}$ increases and reaches values of the order of j. At sufficiently large j and at (12) the state $|\tilde{J}_-\rangle\langle\tilde{J}_-|$ tends asymptotically to the coherent state $|\theta_0, \varphi_0\rangle\langle\theta_0, \varphi_0|$, where $\varphi_0 = \pi/2$, $\cos\theta_0 = [1 - (\bar{v}/j)^2]^{1/2}$; this state corresponds to the classical solution of (1).[13]

The mean values $\langle \hat{J}_z \rangle$, calculated with $\hat{\rho} = |\tilde{J}_-\rangle\langle\tilde{J}_-|$ and $\hat{\rho} = |\theta_0, \varphi_0\rangle\langle\theta_0, \varphi_0|$, as well as the values of $\alpha = |c_j\bar{v}|^2$, shown in Fig. 1 as functions of $\beta = \bar{v}/j$, confirm fully the foregoing estimates.

When $\bar{v}$ exceeds $2j/e$, the solution (7) no longer holds. As noted above, at $\varepsilon = 0$ an exact solution of the stationary equation (1) is possible, and there is no need to resort for this purpose to the numerous methods already used to solve (1). The exact solution is obtained from the general form of Eq. (1). In fact, according to (6), in the stationary regime the operators $\hat{\rho}\hat{J}_-$ and $\hat{J}_-\hat{\rho}$ should commute respectively with the operators $\hat{J}_-$ and $\hat{J}_+$, i.e., the following relation should hold:
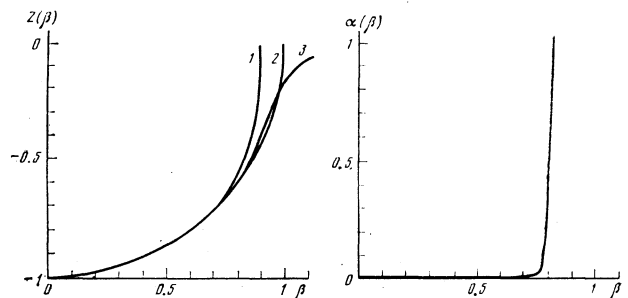
$$\hat{J}_-\hat{\rho} = f(J_+), \tag{17}$$

FIG. 1. Dependences of the quantities $Z = \langle \hat{J}_z \rangle /j$ and $\alpha = |c_j\bar{v}|^2$ on $\beta = \bar{v}/j$ at $j = 20$. Curve 1 was calculated with $\hat{\rho} = |\tilde{J}_-\rangle\langle\tilde{J}_-|$, curve 2 with $\hat{\rho} = |\theta_0, \varphi_0\rangle\langle\theta_0, \varphi_0|$, and curve 3 with the exact value of $\hat{\rho}$.

where f is an arbitrary function. Since the operator $\hat{\rho}$ is Hermitian, we get from (17)

$$\hat{\rho}=\bar{A}^{-1}(\hat{J}_-)^{-1}(\hat{J}_+)^{-1}. \qquad (18)$$

Since the operators

$$(1-\hat{B})^{-1}=1+\sum_{n=1}^{\infty}\hat{B}^n$$

are defined only for operators $\hat{B}$ that have eigenvalues,[19] while the operators $\hat{J}_-,\hat{J}_+$ have none, an additional condition must be introduced to be able to use (18). Taking this condition to be that the density matrix tend to the ground state as $\bar{v}\to 0$,

$$\hat{\rho}_{\bar{v}\to 0}=[1/(2j)!]^2(J_-)^{2j}(J_+)^{2j}$$

and taking into account the relations

$$(J_-)^{2j+1}=(J_+)^{2j+1}=0,$$

we obtain ultimately

$$\hat{\rho}=\bar{A}^{-1}\sum_{k=0}^{2j}\sum_{l=0}^{2j}(-i\bar{v})^{2j-k}(i\bar{v})^{2j-l}(J_-)^k(J_+)^l, \qquad (19)$$

where

$$\bar{A}=\mathrm{Sp}(\hat{J}_-)^{-1}(\hat{J}_+)^{-1}=\sum_{m=0}^{2j}\sum_{n=0}^{m}\alpha_{mn}\bar{v}^{2(2j-n)}, \qquad (19a)$$

$$\alpha_{mn}=[(2j-m+n)!m!]/[(m-n)!(2j-m)!].$$

It follows from (19) that as $\bar{v}\to\infty$ saturation $\hat{\rho}_\infty=\hat{1}$ sets in.

From the plots of the mean values

$$Z(\beta)=\frac{\langle\hat{J}_z\rangle}{j}=\frac{1}{j\bar{A}}\sum_{m=0}^{2j}\sum_{n=0}^{m}(j-m)\alpha_{mn}\bar{v}^{2(2j-n)}, \qquad (20a)$$

$$Y(\beta)=\frac{\langle\hat{J}_y\rangle}{j}=\frac{\bar{v}}{j\bar{A}}\sum_{m=1}^{2j}\sum_{n=1}^{m}\alpha_{mn}\bar{v}^{2(2j-n)}, \qquad (20b)$$

$$Z_1(\beta)=\frac{\langle\hat{J}_z^2\rangle}{j^2}=\frac{1}{j^2\bar{A}}\sum_{m=0}^{2j}\sum_{n=0}^{m}(j-m)^2\alpha_{mn}\bar{v}^{2(2j-n)}, \qquad (20c)$$

$$Z_2(\beta)=Z_1-Z^2, \qquad (20d)$$

shown in Fig. 2 it is seen that at $\beta=\bar{v}/j=1$, in the case of large j, a rather abrupt transition takes place from the region $\beta<1$, in which the system exhibits coherent (classical) properties $Y\approx\beta$, $Z_2\approx 0$, into the region $\beta>1$, in which quantum effects are significant: $Z_2\ne 0$, the average induced dipole moment Y is not proportional to the amplitude β of the incident radiation. In the limit as $j\to\infty$, but $\beta=\bar{v}/j$ is finite, the system undergoes at $\beta=1$ a near-second-order transition. The asymptotic values of the functions (20) are given in this limit by the relations

$$\bar{Z}(\beta)=\begin{cases}(1-\beta^2)^{1/2}, & \beta<1\\ 0, & \beta>1\end{cases}; \qquad (20'a)$$

$$\bar{Y}(\beta)=\begin{cases}\beta, & \beta<1\\ \beta-(1-\beta^2)^{1/2}\arcsin(1/\beta), & \beta>1\end{cases}; \qquad (20'b)$$

$$\bar{Z}_1(\beta)=\begin{cases}1-\beta^2, & \beta<1\\ 1-\beta\bar{Y}(\beta), & \beta>1\end{cases}; \qquad (20'c)$$

$$\bar{Z}_2(\beta)=\begin{cases}0, & \beta<1\\ \bar{Z}_1(\beta), & \beta>1\end{cases}. \qquad (20'd)$$

It must be noted here that the asymptotic values of Y, Z, and Z at $\beta\to 1$ can be obtained with the aid of the meth-
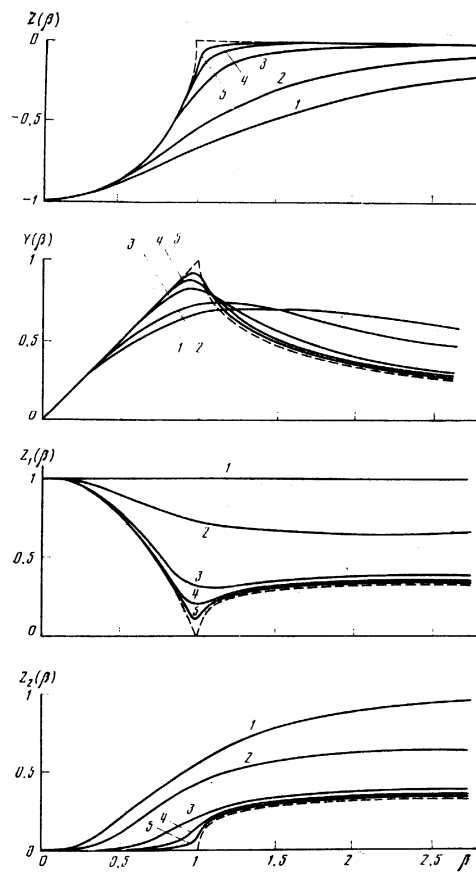

FIG. 2. The functions $Z(\beta)$, $Y(\beta)$, $Z_1(\beta)$ and $Z_2(\beta)$ at $j=0.5$ (1), 1 (2), 5 (3), 15 (4), 50 (5). The dashed curves are the asymptotic values of the functions as $j\to\infty$.

od of Ref. 20 by averaging the classical solutions obtained in Ref. 13. This is in fact how the value of $\bar{Y}(\beta)$ was obtained in Ref. 14. From Fig. 2 and relation (4) it follows also that unlike in monatomic resonant fluorescence the dependence of the integrated intensity $\langle\hat{J}_+\hat{J}_-\rangle$ of cooperative resonant fluorescence on the power of the exciting radiation is not monotonic. Even at relatively small N the function $\langle\hat{J}_+\hat{J}_-\rangle$ has a maximum at $\beta=1$.

The spontaneous-emission spectrum is determined by the Fourier transform of the two-dimensional mean value $\langle\hat{J}_+^{(t)}\hat{J}_-^{(t+\tau)}\rangle$. In the basis of the eigenstates of the operator $\hat{J}_z$ we have

$$\langle\hat{J}_+^{(t)}\hat{J}_-^{(t+\tau)}\rangle$$

$$=\sum_{k,l=-j}^{j}(\lambda_k\lambda_l)^{1/2}\sum_{n=-j}^{j}\begin{cases}D_{k-1,k}^{l-1,n(\tau)}\rho_{ln}^{(t+\tau)}, & \tau<0\\ D_{l,l-1}^{n,k-1(\tau)}\rho_{nk}^{(t)}, & \tau>0\end{cases} \qquad (21)$$

where $\lambda_k=(j+k)(j-k+1)$, and $D_{ij}^{kl\,(\tau)}$ is the Green's function of Eqs. (1) $(D_{ij}^{kl\,(0)}=\delta_{ik}\delta_{jl})$. It is impossible to solve the time-dependent Eq. (1) by the method described above. However, using perturbation theory, we can obtain approximate expressions for the spectrum. Thus, in the $(2j+1)$st order in the intensity of the exciting radiation, a noncoherent component with a Lorentz contour appears in the spectrum and has a half-wdith γj. With increasing radiation intensity,

owing to the interference effect,[21] this component ceases to have a Lorentz shape and its width decreases. At sufficiently large $\bar{v}$ the Stark shift of the levels gives rise to sideband components with frequencies $\omega_0 \pm 2v$. There are also components at the frequencies $\omega_0 \pm 2vk$ ($k = 2, 3, \ldots, N$). Their intensity, however, is much lower than that of the components $\omega_0 \pm 2v$, on the wings of which they in fact appear. In the limit $\bar{v}/j \gg 1$ the spectrum goes over into a single-particle spectrum: three Lorentz components ω_0 and $\omega_0 \pm 2v$ with half-widths $\frac{1}{2}\gamma$ and $(3/4)\gamma$ and with intensity ratio $2:1$. The only difference lies in the intensity of the spectrum, which is $\frac{1}{3}N(N+2)$ times larger for the N-particle problem than for the single-particle problem.

In the case of cooperative resonance fluorescence, the intensity correlation functions undergo larger changes. These changes will be considered below jointly with the case of widely spaced components.

3. We consider the influence of the frequency detuning ε on the cooperative resonant fluorescence, using an example in which the spectral components are quite widely spaced. A numerical formulation of this condition will be presented below.

It is well known that the field of the scattered radiation can be expressed in terms of the Heisenberg operators $\hat{J}_{\pm}^{(t)}$. Carrying out the unitary rotation transformation

$$\hat{A}' = \exp(-i\theta J_y)\hat{A}\exp(i\theta J_y), \tag{22}$$

$$J_z' = \cos\theta J_z + \sin\theta J_x \equiv \Omega^{-1}\hat{V}, \tag{22a}$$

$$J_x' = -\sin\theta J_z + \cos\theta J_x, \tag{22b}$$

where $\sin\theta = 2v/\Omega$, $\cos\theta = \varepsilon/\Omega$, $\Omega^2 = 4v^2 + \varepsilon^2$, we obtain in the interaction representation with respect to V

$$J_{int+}^{(t)} = e^{i\hat{V}t}\hat{J}_+ e^{-i\hat{V}t}e^{i\omega_0 t} = \hat{S}_+^0 e^{i\omega_0 t} + \hat{S}_+^+ e^{i(\omega_0+\Omega)t} + \hat{S}_+^- e^{i(\omega_0-\Omega)t}, \tag{23}$$

where

$$\hat{S}_+^0 = \sin\theta J_z', \qquad \hat{S}_+^{\pm} = (\cos\theta \pm 1)J_{\pm}'/2. \tag{23a}$$

The expansion (23) is valid also in the Heisenberg representation. In this case, however, the operators $\hat{S}^\alpha$ depend on the time. If the Rabi frequency Ω is much higher than the rate of change of the operators $\hat{S}^\alpha$ as a result of relaxation, i.e.,

$$\Omega \gg \gamma j, \tag{24}$$

then each of the operators $\hat{S}^\alpha$ can be regarded as a source of a field at the corresponding frequency. The intensity of the α component is proportional to the mean value $\langle \hat{S}_+^\alpha \hat{S}_-^\alpha \rangle$, and the spectrum is proportional to the autocorrelation function $\langle \hat{S}_+^{\alpha(t)} \hat{S}_-^{\alpha(t+\tau)} \rangle$. To find these quantities as well as the correlation function of the intensities of the scattered radiation, we must solve the time-dependent Eq. (1). In the interaction representation in $\hat{V}$, and if (24) is satisfied, we can average in (1) over the system's fast motion, whose characteristic time is Ω^{-1}. Taking (23) and (23a) into account we obtain

$$\frac{d\hat{\rho}}{dt} = \frac{\gamma}{2}\left\{\sin^2\theta([J_z', \rho J_z'] + [J_z'\rho, J_z']) + \left(\frac{1-\cos\theta}{2}\right)^2 ([J_+', \hat{\rho}J_-'] \right.$$

$$\left. + [J_+'\hat{\rho}, J_-']) + \left(\frac{1+\cos\theta}{2}\right)^2 ([J_-', \hat{\rho}J_+'] + [J_-'\hat{\rho}, J_+']) \right\}. \tag{25}$$

The obtained equation is fully equivalent to the equation for the relaxation of a spin j in contact with a thermostat[21] (with the exception of the terms proportional to $\sin^2\theta$). The role of the Boltzmann factor $\xi = e^{-\hbar\omega_0/kT}$ is assumed here by the parameter

$$\xi = (1-\cos\theta)^2/(1+\cos\theta)^2. \tag{26}$$

We note that the terms proportional to $\sin^2\theta$ correspond to the terms that describe the adiabatic broadening of the transitions in ordinary relaxation theory.[22] In fact, if we introduce the probability of the transitions between the eigenstates of the operator $\hat{J}_z$

$$d_{m+1, m} = \gamma\lambda_{m+1}(1+\cos\theta)^2/4, \quad d_{m-1, m} = \gamma\lambda_m(1-\cos\theta)^2/4,$$

then the total width of the mn transition is

$$\gamma_{mn} = \Delta_{mn} + \sum_k (d_{mk}+d_{km})/2,$$

while $\Delta_{mn} = \gamma(m-n)^2\sin^2\theta/2$ and describes the adiabatic broadening of the mn transition.

By virtue of the analogy noted above, the stationary solution of (25) is

$$\hat{\rho} = Q^{-1}\exp(-\mu J_z'), \tag{27}$$

where $e^{-\mu} = \xi$, and

$$Q = \text{Sp}\exp(-\mu J_z') = \text{sh}[(2j+1)\mu/2]/\text{sh}(\mu/2). \tag{28}$$

The average powers of the operator J_z' are given by the relation

$$\langle (J_z')^n \rangle = (-1)^n Q^{-1}d^n Q/d\mu^n. \tag{29}$$

The dependences of the mean values

$$Z(\beta) = j^{-1}\langle J_z \rangle = j^{-1}\cos\theta\langle J_z' \rangle,$$

$$X(\beta) = j^{-1}\langle J_x \rangle = j^{-1}\sin\theta\langle J_z' \rangle,$$

$$Z_1(\beta) = j^{-2}\langle J_z^2 \rangle$$

$$= (2j)^{-1}(j+1)\sin^2\theta + (\cos^2\theta - \frac{1}{2}\sin^2\theta)j^{-2}\langle (J_z')^2 \rangle$$

on the parameters $\bar{\beta} = |2v/\varepsilon j|$, calculated on the basis of the obtained expressions, are shown for various j in Fig. 3. The maximum of the induced dipole moment X is reached at $|2v/\varepsilon| \sim 1$, in exact agreement with the condition $\beta \sim 1$ at $\varepsilon = 0$ as a consequence of (24). At the same value of $|2v/\varepsilon|$, the effects connected with saturation become significant.

It follows from a comparison of Figs. 2 and 3 that the influence of the detuning on the collective emission manifests itself strongly at small values of v. Thus, whereas at $\varepsilon = 0$ the spectrum contains only the coherent component up to the $2j$-th order in the intensity of the incident radiation, at $\varepsilon \neq 0$ the noncoherent components at the frequencies $2\omega_0 - \omega$ and ω manifest themselves in second order. In the general case the intensities of the three components of the spectrum are given by the equations

$$I_0 = \hbar\omega_0^4(\gamma/\omega)^3\sin^2\theta\langle (J_z')^2 \rangle, \tag{30a}$$

$$I_{\pm} = \hbar(\omega_0 \pm \Omega)^4\left(\frac{\gamma}{\omega}\right)^3 \frac{(1+\cos\theta)^2}{4}[j(j+1) - \langle (J_z')^2 \rangle + \langle J_z' \rangle]. \tag{30b}$$

The intensity of the coherent component is

$$I_0^c = \hbar\omega_0^4(\gamma/\omega)^3\sin^2\theta\langle J_z' \rangle^2. \tag{30c}$$
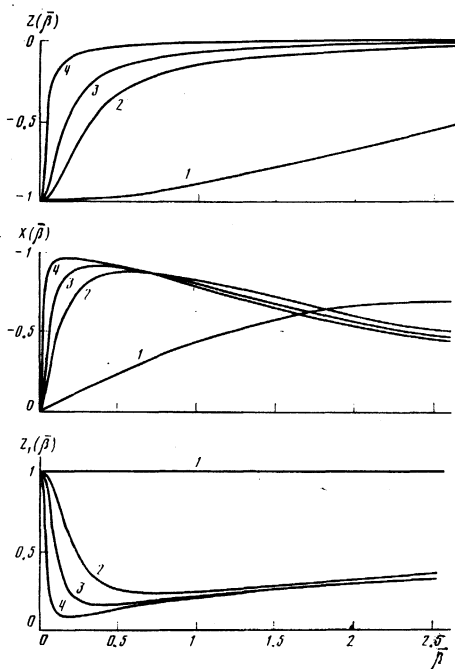
According to these formulas, the intensity of the central

FIG. 3. Dependences of Z, X, and Z_1 on $\bar\beta = |2\nu/\varepsilon j|$ at $j = 0.5$ (1), 5 (2), 10 (3), 40 (4).

component increases with increasing v, reaches a maximum at $|2v/\varepsilon| \sim 1$, and then decreases to a value $\hbar\omega_0^4(\gamma/\omega)^3 j(j+1)/3$. The intensities of the sideband components increase monotonically with increasing v, and reach in the saturation regime values $\hbar(\omega_0 \pm \Omega)^4 \times (\gamma/\omega)^3 j(j+1)/16$. As to the spectrum of these components, using relation (21) and perturbation theory, it is easy to show that at small $|2v/\varepsilon|$ (in second order in $|2v/\varepsilon|$) the sideband components are Lorentzian with half-widths $\gamma j/2$. The central noncoherent component that appears in third order is also Lorentzian; its half-width is γj. As a result of the interference effect they become narrower and go over in the saturation limit into the ordinary components of the one-particle problem. As to the multiple components $\omega_0 \pm k\Omega$, they are absent in the approximation given by (24).

We consider now the correlations of the intensities of the scattered radiation. According to (23), the expression for the intensity correlations consists in the approximation considered of five components: central, two singly shifted by frequencies $\pm\Omega$ and two with double the shift at $\pm 2\Omega$:

$$G^{(2,2)}(\tau) = \langle \hat J_+ \hat J_+^{\prime(\tau)} \hat J_-^{\prime(\tau)} \hat J_- \rangle = G_0(\tau) + 2G_1(\tau)\cos(\Omega\tau) + 2G_2(\tau)\cos(2\Omega\tau),$$

$$G_0(\tau) = [(1 + \cos\theta)/2]^2 \{16\,\xi\,\langle \hat J_z'\hat J_z'^{(\tau)}\hat J_z'^{(\tau)}\hat J_z'\rangle + 4\xi^{1/2}(\langle \hat J_z'\hat J_+'^{(\tau)}\hat J_-'^{(\tau)}\hat J_z'\rangle \quad (31)$$

$$+ \langle \hat J_+'\hat J_z'^{(\tau)}\hat J_z'^{(\tau)}\hat J_-'\rangle) + 4\xi^{1/2}(\langle \hat J_z'\hat J_z'^{(\tau)}\hat J_+'^{(\tau)}\hat J_z'\rangle + \langle \hat J_-'\hat J_z'^{(\tau)}\hat J_z'^{(\tau)}\hat J_+'\rangle)$$

$$+ \xi(\langle \hat J_+'\hat J_-'^{(\tau)}\hat J_+'^{(\tau)}\hat J_-'\rangle + \langle \hat J_-'\hat J_+'^{(\tau)}\hat J_-'^{(\tau)}\hat J_+'\rangle) + \xi^2\langle \hat J_-'\hat J_-'^{(\tau)}\hat J_+'^{(\tau)}\hat J_+'\rangle$$

$$+ \langle \hat J_+'\hat J_+'^{(\tau)}\hat J_-'^{(\tau)}\hat J_-'\rangle\}, \quad (31a)$$

$$G_1(\tau) = \sin^2\theta\{[(1 + \cos\theta)/2]^2\langle J_z'J_+'^{(\tau)} J_z'^{(\tau)} J_-'\rangle$$

$$+[(1 - \cos\theta)/2]^2\langle J_-'J_z'^{(\tau)} J_+'^{(\tau)} J_z'\rangle + (\sin\theta/2)^2[\langle J_z'J_z'^{(\tau)} J_+'^{(\tau)} J_-'\rangle$$

$$+\langle J_-'J_+'^{(\tau)} J_z'^{(\tau)} J_z'\rangle]\}, \quad (31b)$$

$$G_2(\tau) = (\sin\theta/2)^4\langle J_-'J_-'^{(\tau)} J_+'^{(\tau)} J_+'\rangle. \quad (31c)$$

The function $G_i(\tau)$ is expressed in terms of the

Green's matrix of Eq. (25) in a rather complicated manner. Without investigating the entire dependence on τ, we note a number of general aspects.

First is that the spectrum of the intensity correlations of the cooperative fluorescence contains components at the frequencies $\pm 2\Omega$. As follows from (31c), for monatomic fluorescence ($j = \frac{1}{2}$), by virtue of the relation $(\hat J_+)^{2j+1} = 0$, this component is absent. Therefore the presence of the 2Ω component in the spectrum of the intensity fluctuations can serve as a basis for the experimental observation of the cooperative character of the scattering of the experimentally observed cooperative character of the light scattering. This fact has also been noted in Ref. 18 for the particular case $\varepsilon = 0, \bar v \to \infty$. It must be noted here that the onset of the 2Ω components is the result of correlations of the once-shifted components of the spectrum with one another. These correlations are described by the functions $\langle S_+^\alpha S_+^{\alpha\,(\tau)} S_-^{\alpha\,(\tau)} S_-^\alpha \rangle$, $\alpha = \pm$. In the case of monatomic resonant fluorescence they are equal to zero.[5]

Of particular interest is the behavior of the functions $G_i(\tau)$ and $G^{(2,2)}(\tau)$ as $\tau \to 0$, since the function $G^{(2,2)}$ vanishes for monatomic fluorescence at $\tau = 0$ (the photon antibunching effect). Using relations (4), (28), and (29) we can easily show that $G^{(2,2)}(0) \ne 0$ at $j \ne \frac{1}{2}$ and always $G_0(0) \geqslant 0$, $G_2(0) \geqslant 0$, $G_1(0) \leqslant 0$. The nonvanishing of $G^{(2,2)}(0)$ is the result of the simultaneous emission of two or several photons by the system of atoms. At low pump intensities ($\xi \ll 1$) we have

$$G_0(0) \approx 4j^2(4j^2+1)\xi, \quad G_1(0) \approx -8j^3\xi,$$
$$G_2(0) \approx 4j(2j-1)\xi^2. \quad (32)$$

It follows from (32) that at small ξ ($|2v/\varepsilon| \ll 1$) the contributions to $G^{(2,2)}(0)$ are made by the central and once-shifted components. The most substantial in the central component is the coherent component, since $\langle \hat J_+ \hat J_- \rangle \approx 16j^4\xi$. The normalized correlation function of the intensities is in this approximation

$$g(0) = G^{(2,2)}(0)/\langle \hat J_+ \hat J_- \rangle \approx 1 - 1/j + 1/4j^2, \quad (33)$$

whereas at $\varepsilon = 0$ and $\bar v/j \ll 1$ it is equal to unity up to the scattered-light intensity raised to the $2j$ power. Thus, the presence of frequency detuning leads to loss of coherence of the scattered light.

With increasing intensity of the exciting radiation, the functions $G_i(0)/\langle J_+ J_- \rangle^2$ increase monotonically. In the limit as $\xi \to 1$ we have

$$\bar G_0(0) = {}^1/_{60}j(j+1)[28j(j+1)-1],$$
$$\bar G_1(0) = -{}^1/_{6}j(j+1), \quad (34)$$
$$\bar G_2(0) = {}^1/_{120}j(j+1)[4j(j+1)-3].$$

It follows from (34) that in this limit, at large j, the largest contribution to the total correlation function $G^{(2,2)}(0)$ of the intensities is made by the central and the two twice-shifted components. The normalized correlation function is equal to

$$\bar g(0) = {}^3/_{10}[4j(j+1)-3]/j(j+1). \quad (35)$$

Since $G^{(2,2)}(\tau) \to \langle \hat J_+ \hat J_- \rangle^2$ as $\tau \to \infty$, the quantity $g(0)$ characterizes the behavior of the function $G^{(2,2)}(\tau)$ in the mean. From (33) and it follows that at low intensities of the exciting radiation $G^{(2,2)}(\tau)$ is a function that

increases on the average at any τ, since $g(0) < 1$. As $\xi \to 1$ we have $\bar{g}(0) < 1$ at $j = \frac{1}{2}$, 1, and 2 and $\bar{g}(0) > 1$ at $j \geq 2$.

In addition to estimating the behavior of the function $G^{(2,2)}(\tau)$ in the mean, it is of interest to consider the time dependence of $G^{(2,2)}(\tau)$ at small τ. This can be done by representing the formal solution of (1) in the form $\hat{\rho}(\tau) = \exp(\mathscr{L}\tau)\hat{\rho}(0)$, where $\mathscr{L}$ is the Liouville operator of Eq. (1). We then have for the first two coefficients of the expansion of $G^{(2,2)}(\tau)$ in powers of τ

$$k_1 = dG^{(2,2)}(\tau)/d\tau|_{\tau=0} = \mathrm{Sp}\,(J_+ J_- \mathscr{L}(J_- \hat{\rho} J_+)),$$

$$k_2 = d^2 G^{(2,2)}(\tau)/d\tau^2|_{\tau=0} = \mathrm{Sp}\,(J_+ J_- \mathscr{L}(\mathscr{L}(J_- \hat{\rho} J_+))). \tag{36}$$

Using the solution (19) of Eq. (1) at $\varepsilon = 0$, it is easy to prove that in the stationary regime

$$k_1 = 0, \quad k_2 = -\left(\frac{\gamma}{2}\right)^2 \frac{\bar{v}^{2(2j+1)}}{\bar{A}} \frac{4}{15} j(j+1)\,[j(j+1)-2]. \tag{37}$$

It follows from the last relations that at $j > 1$ the intensity-correlation function is at small τ a (quadratically) decreasing function of the time delay, whereas at $j = \frac{1}{2}$ and 1 it increases quadratically and cubically with time, respectively. Thus, at $j > 1$ there is no photon antibunching. At high excitation intensity, the absence of this effect is due to the existence of the component at the double-shifted frequency in the intensity correlation spectrum. In fact, according to (31) and (34) in this case $k_2 = -2(\bar{G}_1 + 4\bar{G}_2)$.

4. The main singularities of resonant fluorescence, when account is taken of the collective character of the emission of light by the system of atoms, reduces to the following:

a) With increasing number of radiation-scattering atoms, an increase takes place in the coherence region (the region in which the scattered radiation is fully coherent) with respect to $\bar{v}$, reaching at $\varepsilon = 0$ values on the order of the number of the radiating atoms. In the limit as $j \to \infty$, at $\beta = \bar{v}/j = 1$ a transition analogous to a second-order phase transition takes place from the coherence region into the region where the quantum properties are significant.

b) The emission spectrum at $\varepsilon = 0$, up to $2j$-th order in the intensity of the exciting radiation, consists of a δ-like coherent component. In the $(2j+1)st$ order a noncoherent component appears already in second order. With increasing excitation intensity, the spectrum is transformed and goes over in the high-intensity limit into the spectrum of monatomic fluorescence, but with an integrated intensity $(4/3)j(j+1)$ times larger. The integrated intensity of the cooperative resonant fluorescence is not a monotonic function of the exciting radiation: at $\varepsilon = 0$ there is a maximum in the region $\beta = 1$.

c) The correlation of the intensities of the radiation scattered by a system of particles differs quite substantially from the case of scattering by a single atom. Above all, the collective character of the radiation manifests itself in the fact that when the spectrum components are widely spaced ($\Omega \gg \gamma j$) the spectrum of the intensity correlations has a component of frequency 2Ω, corresponding to monatomic fluorescence. In the saturation regime, the intensity of this component prevails over the intensity of the component at the frequency 2Ω. In addition, at zero time delay ($\tau = 0$) the intensity correlation function $G^{(2,2)}(\tau)$ is not equal to zero, and at small τ and when the number of scattering particles larger than two, the function $G^{(2,2)}(\tau)$ is a decreasing function of the delay. In this case the photon antibunching effect, which takes place for monatomic fluorescence, is absent.

In conclusion, notice should be taken of the considerable influence exerted on the spectrum of the resonant fluorescence by the direct interaction between the atoms. Within the framework of the model considered in this article, it can be taken into account by introducing in (1) a term $i[\hat{Q}, \hat{\rho}]$, where

$$Q = \sum_{\alpha \neq \beta} \sum \,[\varepsilon_q^{\alpha\beta}(J_z^\alpha + J_z^\beta) - 2q^{\alpha\beta}(J_z^\alpha + J_z^\beta)^2].$$

It is easy to show that this interaction leads, in particular, to an increase of the number of spectral components, and in the saturation regime the spectrum does not become monatomic. Thus, at $j = 1$ the resonant-fluorescence spectrum at $j = 1$ thus consists in the general case of seven components.[10]

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Note added in proof (29 April 1980). When this article was in press, the author learned of the paper by R. R. Puri and S. V. Lawande [Phys. Lett. 72A, 200 (1979)], in which the method of coherent states was used to obtain for Eq. (1) a solution that coincides with solution (19) of the present article.

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Nonlinear processes in vibrational–translational relaxation of a gas of strongly excited molecules

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An analysis is made of the characteristics of vibrational relaxation processes in strongly excited molecules. An important class of new phenomena occurring under these conditions are the nonlinear (depending on the rate of excitation) vibrational relaxation processes. Under strong excitation conditions we can expect the appearance of new relaxation channels (including stimulated processes); another manifestation of nonlinearity is the dependence of the relaxation kinetics on the rate of excitation. The results are reported of experimental and theoretical investigations of nonlinear processes in a vibrational–translational (VT) relaxation in molecular gases CD_4 and SF_6 excited resonantly by laser radiation. Nonexponential VT relaxation processes in strongly excited molecules are characterized by an "instantaneous" relaxation time τ_{VT} dependent on the initial vibrational temperature $T_v(0)$ and on the time t from the moment of excitation: $\tau_{VT} = \tau_{VT}[T_v(0), t]$. The experimental results on the behavior of τ_{VT} of pure gases and mixtures have made it possible to determine the nonlinear mechanisms. It is shown that the contribution to the observed effects is due to the anharmonicity of the vibrations and also due to an increase in the average translational energy of the gas. In the case of the CD_4 molecule these qualitative considerations are supported by a detailed quantitative calculation. In the case of a two-mode model of the vibrations in CD_4 the results of a calculation of "weakly nonlinear" relaxation kinetics agree to within at least 20% with the experimental data. A simple one-mode model may be used for relatively rough estimates in the case of polyatomic molecules. The nonlinear effects can be very strong also in the case of other intermolecular and intramolecular relaxation processes.

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INTRODUCTION

1. Investigation of the mechanisms of transformation of the optical excitation energy by molecules and molecular complexes is undoubtedly one of the central problems in the physics of resonant interaction of laser radiation with matter. Many important results have been obtained in the last decade. Various new experimental methods have been proposed, ways of dissipation of the vibrational excitation energy in gases and liquids have been studied, rates of energy conversion have been determined, and laws governing the distribution of energy between vibrational modes in polyatomic molecules have been investigated (see, for example, the reviews in Refs. 1–5).

One of the interesting aspects of this new stage of research on vibrational relaxation is related to the feasibility of investigating the decay of strongly excited states. Many-photon dissociation, including isotopically selective dissociation of many polyatomic molecules has been achieved using lasers emitting far and near infrared radiation.[6,7] It is natural to expect that in the case of such strong excitation there should be nonlinear vibrational relaxation effects, i.e., the nature of relaxation processes may depend on the rate of excitation. In the case of highly excited systems we can expect new relaxation processes and this is true of intermolecular as well as intramolecular relaxation. On the other hand, the energy transfer rates (characteristic relaxation times) for channels already known may depend on the degree of excitation.

2. These problems are now attracting increasing at-