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COLLECTIVE RESONANCE RAMAN SCATTERING OF AN INTENSE LASER FIELD WITH PHASE AND AMPLITUDE FLUCTUATIONS

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The resonance Raman scattering from a system of N three-level atoms that are driven by a strong laser field with the phase and amplitude fluctuations is considered. The exact on-resonance steady-state solution to the atomic density matrix is obtained. The collective properties of the fluorescent spectrum for both the Rayleigh-type as well as the Raman-type processes are discussed. The collective narrowing of the one-atom spectral lines is predicted.

1. Introduction

It is known[1,2]) that the resonant Raman scattering of an intense laser field by a three-level atomic system is quite different from the ordinary weak-field Raman effect. For very intense laser fields, when both allowed atomic transitions are saturated, there is no clear separation between Rayleigh-type and Raman-type processes. This is because of the modifications of the atomic levels due to the dynamic Stark effect, the most spectacular explanation of which can be given in the "dressed atom" picture[1]). The finite bandwidth of the exciting laser field due to phase and/or amplitude fluctuations can considerably affect the results, as it has recently been shown for optical double resonance[3]). A number of other effects related to the interaction of a three-level atom with resonant laser fields and extensive literature on the subject can be found in ref. 4.

On the other hand, a lot of work has been done to explain the collective properties of many two-level atoms interacting with a resonant laser field[5–10]). It would be interesting to know how the properties of an individual three-level atom interacting with laser fields are modified when the number of atoms

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becomes large. Some recent publications deal with such collective effects in double optical resonance[11]) and the resonant Raman scattering[12,13]).

In this paper, we consider the effects of the driving field fluctuations on the spectrum in the collective resonant Raman process. We use the quantum mechanical master equation approach[14]) and secular approximation[6,13]) to eliminate the rapidly oscillating terms. The theory of multiplicative stochastic processes[15]) is used to obtain the equation for the density matrix averaged over the phase and/or amplitude fluctuations of the exciting field. We assume here that the phase fluctuations are described by a Wiener–Levy process[16,17]) whereas the amplitude fluctuations are described by a nonwhite Gaussion process[18,19]). It is shown that even for fluctuating laser fields an exact steady-state solution of the master equation can be easily obtained if the laser field is tuned to the resonance with the atomic transition. All collective steady-state characteristics of the system can thus be derived with the use of this solution. The equations describing the time evolution of the one-time atomic expectation values averaged over the ensembles of the phase and amplitude fluctuations can also be obtained from the master equation. To solve these equations we have applied a decorrelation scheme that allows for closing the system of equations, and to calculate the two-time correlation functions the quantum regressian theorem is invoked. We have derived explicit analytical formulas for the field correlation functions of the scattered light that explain the effects of the laser field fluctuations. The collective properties of the scattered light are discussed showing a possibility of collective narrowing of the one-atom spectral widths.

2. Master equation and steady-state averages

We consider a system of N three-level atoms confined to a region small compared to the wavelengths of all relevant radiation modes (the Dicke model) interacting with a driving laser field of frequency ω_L and with the vacuum of all other modes. A schematic diagram of atomic energy levels is shown in fig. 1. The ground state $|1\rangle$ is coupled to the state $|2\rangle$ by the strong,

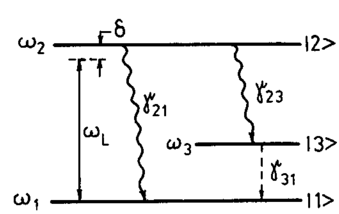

Fig. 1. Schematic diagram of energy levels and possible transitions for the three-level atom considered in this paper.

resonant laser field, and there is a spontaneous transition from the level $|2\rangle$ to $|3\rangle$ (Stokes line). The dipole transition between the levels $|3\rangle$ and $|1\rangle$ is forbidden due to parity considerations, and we introduce a nonradiative relaxation mechanism (which we do not specify) that makes the transition $|3\rangle \to |1\rangle$ possible. The nonzero transition rate for the transition $|3\rangle \to |1\rangle$ is important in order to have a nontrivial steady-state solution for the density matrix.

On treating the exciting laser field classically and making standard (Born and Markov) approximations to describe the system–reservoir couplings, one obtains a master equation for the reduced density operator ρ of the atomic system alone in the following form[14]) ($\hbar = 1$ units are used):

$$\begin{aligned}\frac{\partial \rho}{\partial t} = & -\mathrm{i}\frac{d}{2}[E^*(t)J_{21} + E(t)J_{12}, \rho] - \mathrm{i}\frac{\delta}{2}[J_{22} - J_{11}, \rho] - \mathrm{i}\Omega_3[J_{33}, \rho] \\ & - \gamma_{21}(J_{21}J_{12}\rho - J_{12}\rho J_{21} + \mathrm{h.c.}) - \gamma_{23}(J_{23}J_{32}\rho - J_{32}\rho J_{23} + \mathrm{h.c.}) \\ & - \gamma_{31}(J_{31}J_{13}\rho - J_{13}\rho J_{31} + \mathrm{h.c.}), \end{aligned} \quad (1)$$

where $\Omega_3 = \omega_{23} - \omega_{21}/2$, $2\gamma_{21}$ and $2\gamma_{23}$ are the single-atom spontaneous emission rates for the transitions $|2\rangle \to |1\rangle$ and $|2\rangle \to |3\rangle$, respectively, and $2\gamma_{31}$ is the nonradiative transition rate for the transition $|3\rangle \to |1\rangle$; $\delta = \omega_{21} - \omega_{\mathrm{L}}$ is the detuning of the laser frequency ω_{L} from the atomic transition frequency ω_{21}. The collective atomic operators J_{ij} are defined as

$$J_{ij} = \sum_{k=1}^{N} |i\rangle_{k\,k}\langle j| \quad (i, j = 1, 2, 3)$$

and satisfy the commutation relations

$$[J_{ij}, J_{i'j'}] = J_{ij'}\delta_{i'j} - J_{i'j}\delta_{ij'} .$$

In the master equation (1) the rotating wave approximation has been used and the equation is written in the frame rotating with respect to the laser frequency.

We assume that the laser field is described by

$$E(t) = [E_0 + \Delta E(t)]\, \mathrm{e}^{-\mathrm{i}\varphi(t)}, \quad (2)$$

where E_0 and $\varphi_0 = \varphi(0)$ are the nonstochastic parts of the field amplitude and phase, respectively, while the time-dependent quantities $\Delta E(t)$ and $\varphi(t)$ are stochastic variables describing the amplitude and phase fluctuations of the laser field, respectively.

As is usually done[17–19]), we will model the phase fluctuations by the Wiener–Levy process (phase diffusion model) and the amplitude fluctuations by a nonwhite Gaussian noise. So, we have

$$\frac{\mathrm{d}\varphi(t)}{\mathrm{d}t} = \mu(t) , \tag{3}$$

where $\mu(t)$ is a Gaussian white noise with zero mean value and the correlation function

$$\overline{\mu(t)\mu(t')} = 2\gamma_c \delta(t - t') \tag{4}$$

and

$$\overline{\Delta E(t)} = 0 ,$$
$$\overline{\Delta E(t)\Delta E(t')} = (\Delta E)^2 \, \mathrm{e}^{-\gamma_a |t-t'|} , \tag{5}$$

where $(\Delta E)^2$ is a measure of the amplitude fluctuations, and γ_c and γ_a describe finite bandwidths due to phase and amplitude fluctuations, respectively. The bar is used to denote the average value over an ensemble of phase or amplitude fluctuations. A double bar will be used to denote averaging over both phase and amplitude fluctuations. We treat here the phase and amplitude fluctuations as independent stochastic processes.

To proceed further, we adopt the procedure used by Puri and Lawande[8]) in the calculations of laser fluctuation effects in a system of two-level atoms. We introduce the transformation

$$W_m(t) = \mathrm{e}^{-\mathrm{i}m\varphi(t)} \, \mathrm{e}^{-(\mathrm{i}/2)\varphi(t)(J_{22}-J_{11})} \rho \, \mathrm{e}^{(\mathrm{i}/2)\varphi(t)(J_{22}-J_{11})} \tag{6}$$

into the master equation (1), and obtain the following equation for the transformed quantity:

$$\frac{\mathrm{d}W_m(t)}{\mathrm{d}t} = [L_0 - \mathrm{i}(m + L_1)\dot{\varphi}(t) - \mathrm{i}\epsilon(t)L_2]W_m(t) , \tag{7}$$

where

$$\begin{aligned} L_0 W_m = & -\mathrm{i}\epsilon_0[J_{21} + J_{12}, W_m] - \mathrm{i}\,\frac{\delta}{2}\,[J_{22} - J_{11}, W_m] \\ & -\mathrm{i}\Omega_3[J_{33}, W_m] - \gamma_{21}(J_{21}J_{12}W_m - 2J_{12}W_m J_{21} + W_m J_{21}J_{12}) \\ & -\gamma_{23}(J_{23}J_{32}W_m - 2J_{32}W_m J_{23} + W_m J_{23}J_{32}) \\ & -\gamma_{31}(J_{31}J_{13}W_m - 2J_{13}W_m J_{31} + W_m J_{31}J_{13}) , \end{aligned} \tag{8}$$

$$L_1 W_m = \tfrac{1}{2}[J_{22} - J_{11}, W_m] ,$$

$$L_2 W_m = [J_{21} + J_{12}, W_m]$$

and $\epsilon_0 = \frac{1}{2} d E_0$, $\epsilon(t) = \frac{1}{2} d \Delta E(t)$ with $d = d_{21}$ being the transition dipole moment between the states $|1\rangle$ and $|2\rangle$.

Taking into account the stochastic properties (eqs. (3) and (4)) of the phase fluctuations and applying the theory of multiplicative stochastic processes[15]) one can obtain the master equation for the density matrix averaged over the phase fluctuations, $\bar{W}_m(t)$, which has the form

$$\frac{\mathrm{d}\bar{W}_m(t)}{\mathrm{d}t} = [L_0 - \gamma_c (m + L_1)^2 - \mathrm{i}\epsilon(t) L_2] \bar{W}_m(t) . \tag{9}$$

Since the operator L_2 which is multiplied by the time-dependent coefficient $\epsilon(t)$ does not commute with all other operators in (9), it is not possible to use the theory of multiplicative stochastic processes to obtain the master equation for the density matrix averaged over the amplitude fluctuations in the same fashion as for the phase fluctuations. We thus restrict our considerations to strong laser fields only. To make these considerations more transparent we introduce the Schwinger representation for the atomic (angular momentum) operators[20]):

$$J_{ij} = c_i^+ c_j \quad (i, j = 1, 2, 3) , \tag{10}$$

where the operators c_i obey the boson commutation rules $[c_i, c_j^+] = \delta_{ij}$.

After performing the canonical (dressing) transformation

$$\begin{aligned} c_1 &= Q_1 \cos \nu + Q_2 \sin \nu , \\ c_2 &= -Q_1 \sin \nu + Q_2 \cos \nu , \\ c_3 &= Q_3 , \end{aligned} \tag{11}$$

where

$$\mathrm{tg}\, 2\nu = 2\epsilon_0 / \delta ,$$

one can split the Liouville operator appearing in eq. (9) into the slowly varying part and the terms oscillating at frequencies 2Ω and 4Ω, with Ω denoting one half of the Rabi frequency. We assume here that the Rabi frequency is sufficiently large and satisfies the relation

$$\Omega = \tfrac{1}{2}(\delta^2 + 4\epsilon_0^2)^{1/2} \ll N\gamma_{21}; N\gamma_{23}; N\gamma_{31} , \tag{12}$$

but $\Omega \ll \omega_{31}$ and the transition $|3\rangle \to |2\rangle$ is not affected by the laser field. In this case the secular approximation[6,13]) is justified and we retain only the slowly varying part of the Liouville operator. We then have (the prime is used to distinguish the transformed density matrix)

$$\frac{\mathrm{d}\bar{W}'_m(t)}{\mathrm{d}t} = [\mathscr{L}_0 - 2\epsilon(t) \sin \nu \cos \nu \, \mathscr{L}_1]\bar{W}'_m(t) , \tag{13}$$

where

$$\begin{aligned}\mathscr{L}_0\bar{W}'_m(t) = &-\gamma_c m^2 \bar{W}'_m(t) - (\mathrm{i}\Omega + m\gamma_c')[D_3, \bar{W}'_m(t)] \\ &-\mathrm{i}\Omega_3[R_{33}, \bar{W}'_m(t)] - \gamma_0[D_3, [D_3, \bar{W}'_m(t)]] \\ &-\gamma_1[R_{21}, [R_{12}, \bar{W}'_m(t)]] - \gamma_2[R_{12}, [R_{21}, \bar{W}'_m(t)]] \\ &-\gamma_{23} \sin^2\nu \, [R_{13}, [R_{31}, \bar{W}_m'(t)]] - \gamma_{23} \cos^2 \nu \, [R_{23}, [R_{32}, \bar{W}'_m(t)]] \\ &- \gamma_{31} \cos^2 \nu \, [R_{31}, [R_{13}, \bar{W}'_m(t)]] \\ &-\gamma_{31} \sin^2 \nu \, [R_{32}, [R_{23}, \bar{W}'_m(t)]] , \\ \mathscr{L}_1\bar{W}'_m(t) = &[D_3, \bar{W}'_m(t)] .\end{aligned} \tag{14}$$

In eqs. (14) we used the notation

$$\begin{aligned}\gamma'_c &= \gamma_c(\cos^2 \nu - \sin^2 \nu) , \\ \gamma_0 &= \gamma_{21} \sin^2 \nu \cos^2 \nu + \frac{\gamma_c}{4} (\cos^2 \nu - \sin^2 \nu)^2 , \\ \gamma_1 &= \gamma_{21} \cos^4 \nu + \gamma_c \sin^2 \nu \cos^2 \nu , \\ \gamma_2 &= \gamma_{21} \sin^4 \nu + \gamma_c \sin^2 \nu \cos^2 \nu , \\ D_3 &= R_{22} - R_{11}\end{aligned} \tag{15}$$

and the operators $R_{ij} = Q_i^+ Q_j$ are new, dressed atomic operators that satisfy the same commutation relations as the operators J_{ij} (the transformation (11) is canonical).

It is easy to check that the operator $\mathscr{L}_1$ commutes with $\mathscr{L}_0$ and the theory of multiplicative stochastic processes can now be used giving the following master equation for the density matrix averaged over the amplitude fluctuations:

$$\frac{\mathrm{d}}{\mathrm{d}t} \bar{\bar{W}}'_m(t) = [\mathscr{L}_0 + \mathscr{L}_2]\bar{\bar{W}}'_m(t) . \tag{16}$$

The Liouville operator $\mathscr{L}_0$ has the same form as given in (14) while $\mathscr{L}_2$ is given by

$$\mathscr{L}_2\bar{\bar{W}}'_m(t) = [\eta(t) - \eta(0)][D_3, [D_3, \bar{\bar{W}}'_m(t)]] , \tag{17}$$

with

$$\eta(t) = \frac{4\epsilon^2}{\gamma_a} \sin^2 \nu \cos^2 \nu \, e^{-\gamma_a t} , \tag{18}$$

where $\epsilon^2 = \frac{1}{4} d^2 (\Delta E)^2$ is a measure of the Rabi frequency fluctuations.

The master equation (16) can be used to calculate the expectation values of the atomic observables averaged over the phase and amplitude fluctuations. When the laser field is tuned to the resonance with the atomic transition $|1\rangle \rightarrow |2\rangle$ ($\delta = 0$), we have $\sin^2 \nu = \cos^2 \nu = \frac{1}{2}$ and the master equation (16) has an exceptionally simple stationary solution

$$\bar{\bar{W}}_m^{\prime\,(s)} = \begin{cases} 0 & \text{for } m \neq 0 , \\ A^{-1} \sum\limits_{R=0}^{N} X^R \sum\limits_{N_1=0}^{R} |R, N_1\rangle\langle N_1, R| , \end{cases} \tag{19}$$

where $X = \gamma_{31}/\gamma_{23}$ and

$$A = \frac{(N+1)X^{N+2} - (N+2)X^{N+1} + 1}{(X-1)^2} .$$

The states $|R, N_1\rangle$ are the eigenstates of the operators R_{11} (the eigenvalues N_1), $R = R_{11} + R_{22}$ (the eigenvalues R) and the operator $\hat{N} = R_{11} + R_{22} + R_{33}$ with the eigenvalue N being the number of atoms.

The solution (19) allows to calculate all stationary expectation values of the atomic observables. Some of the results that will be needed in our further considerations are given in the appendix. It is interesting to notice, however, that the solution (19) does not depend on the laser field fluctuations parameters.

3. Time-dependent averages

The master equation (16) can be used to describe the time evolution of the expectation values of the atomic observables. The parameter m in the transformed density matrix $\bar{\bar{W}}_m'(t)$ has to be chosen appropriately to the character of the operator the average of which is to be calculated. It is easy to show using the transformation (6) and the commutation rules for the atomic operators that, for example,

$$\mathrm{Tr}\{\overline{\overline{(J_{22} - J_{11})\rho(t)}}\} = \mathrm{Tr}\{(J_{22} - J_{11})\bar{\bar{W}}_0(t)\} ,$$

$$\mathrm{Tr}\{\overline{\overline{J_{21}\rho(t)}}\} = \mathrm{Tr}\{J_{21}\bar{\bar{W}}_1(t)\} ,$$

$$\mathrm{Tr}\{\overline{\overline{J_{23}\rho(t)}}\} = \mathrm{Tr}\{J_{23}\bar{\bar{W}}_{1/2}(t)\} \,. \tag{20}$$

So, knowing the averages calculated with the transformed density matrix one can get the "true" averages by putting for m the appropriate value.

For an arbitrary operator Q, according to (16), we have

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\langle Q\rangle_m = &-\gamma_c m^2\langle Q\rangle_m - (\mathrm{i}\Omega + \gamma_c')\langle[Q, D_3]\rangle_m \\ &-(\gamma_0 + \eta(0) - \eta(t))\langle[D_3, [D_3, Q]]\rangle_m \\ &-\gamma_1\{\langle[Q, R_{21}]R_{12}\rangle_m + \langle R_{21}[R_{12}, Q]\rangle_m\} \\ &-\gamma_2\{\langle[Q, R_{12}]R_{21}\rangle_m + \langle R_{12}[R_{21}, Q]\rangle_m\} \\ &-\gamma_{23}\{\sin^2\nu\,[\langle[Q, R_{13}]R_{31}\rangle_m + \langle R_{13}[R_{31}, Q]\rangle_m] \\ &+\cos^2\nu\,[\langle[Q, R_{23}]R_{32}\rangle_m + \langle R_{23}[R_{32}, Q]\rangle_m]\} \\ &-\gamma_{31}\{\cos^2\nu\,[\langle[Q, R_{31}]R_{13}\rangle_m + \langle R_{31}[R_{13}, Q]\rangle_m] \\ &+\sin^2\nu\,[\langle[Q, R_{32}]R_{23}\rangle_m + \langle R_{32}[R_{23}, Q]\rangle_m]\} \,,\end{aligned} \tag{21}$$

where $\langle Q\rangle_m = \mathrm{Tr}\{Q\bar{\bar{W}}{}'_m(t)\}$. We shall use eq. (21) to find the evolution of atomic observables. Further on, we assume the exact resonance case, $\delta = 0$, in which $\cos^2\nu = \sin^2\nu = \frac{1}{2}$. We then have

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\langle R_{12}\rangle_m = &-[2\mathrm{i}\Omega + \gamma_c m^2 + \gamma_1 + \gamma_2 + \gamma_{23} + 4(\gamma_0 + \eta(0) - \eta(t))]\langle R_{12}\rangle_m \\ &-(\gamma_{23} - \gamma_{31})\langle R_{33}R_{12}\rangle_m \,,\end{aligned} \tag{22}$$

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\langle R_{13}\rangle_m = &-[\mathrm{i}(\Omega_3 + \Omega) + \gamma_c m^2 + \tfrac{1}{2}(\gamma_1 + \gamma_2) \\ &+\tfrac{1}{2}\gamma_{23} + \gamma_{31} + \gamma_0 + \eta(0) - \eta(t)]\langle R_{13}\rangle_m \\ &-\tfrac{1}{4}(\gamma_{23} - \gamma_{31})[\langle\{R_{33} - R_{11}, R_{13}\}\rangle_m - \langle R_{22}R_{13}\rangle_m] \,,\end{aligned} \tag{23}$$

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\langle R_{23}\rangle_m = &-[\mathrm{i}(\Omega_3 - \Omega) + \gamma_c m^2 + \tfrac{1}{2}(\gamma_1 + \gamma_2) \\ &+\tfrac{1}{2}\gamma_{23} + \gamma_{31} + \gamma_0 + \eta(0) - \eta(t)]\langle R_{23}\rangle_m \\ &-\tfrac{1}{4}(\gamma_{23} - \gamma_{31})[\langle\{R_{33} - R_{22}, R_{23}\}\rangle_m - \langle R_{11}R_{23}\rangle_m] \,,\end{aligned} \tag{24}$$

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\langle R_{22} - R_{11}\rangle_m = &-[\gamma_c m^2 + 2(\gamma_1 + \gamma_2) + \gamma_{23}]\langle R_{22} - R_{11}\rangle_m \\ &-(\gamma_{23} - \gamma_{31})\langle R_{33}(R_{22} - R_{11})\rangle_m \,,\end{aligned} \tag{25}$$

where

$$\{A, B\} = AB + BA\,.$$

Eqs. (22)–(25) are so far exact. They contain, however, terms with products of operators, which make them unsolvable. Therefore, some approximations are needed. To deal with the product terms we apply a decorrelation scheme similar to that used by Compagno and Persico[5]). The only difference consists in the fact that we decorrelate symmetrized products of operators (anticommutators). This allows us to preserve one-atom terms unchanged and clearly separate them from the collective terms. The decorrelated operators that do not enter the equations as "proper" variables are replaced by their steady-state averages calculated with the density matrix (19). For example, we assume

$$\langle\{R_{ii}, R_{kl}\}\rangle_m = 2\langle R_{ii}R_{kl}\rangle_m = 2\langle R_{ii}\rangle_s \cdot \langle R_{kl}\rangle\,, \tag{26}$$

where

$$\langle A\rangle_s = \mathrm{Tr}\{A\bar{\bar{W}}_0'^{(s)}\}\,.$$

With such approximations eqs. (22)–(25) have simple exponential solutions with the one-atom and collective damping constants clearly separated. On neglecting the collective part one immediately obtains the one-atom results. Of course, for large numbers of atoms the collective part can dominate over the one-atom part, and the latter has little importance. By using the density matrix (19) one can show that[13]) in the case of large N the factorization (26) yields a small error (with an order of $N^{-1/2}$) in the calculation of the steady-state fluorescent spectrum. The explicit expressions for the collective terms can be obtained with the use of the steady-state averages given in the appendix.

4. Two-time correlation functions

Since the operator $\mathscr{L}_2$ in the master equation (16) commutes with the operator $\mathscr{L}_0$ this equation describes a Markov process despite the time-dependent coefficient $\eta(t)$. Thus, the two-time averages may be derived from the one-time averages by taking advantage of the quantum regression theorem[21]). The spectrum of light spontaneously emitted due to the transitions $|2\rangle \to |1\rangle$ and $|2\rangle \to |3\rangle$ is proportional to the Fourier transforms of the following atomic correlation functions:

$$\begin{aligned} \langle\overline{\overline{J_{21}(\tau)J_{12}}}\rangle_{ss} &= \lim_{t\to\infty} \langle\overline{\overline{J_{21}(t+\tau)J_{12}(t)}}\rangle\,, \\ \langle\overline{\overline{J_{23}(\tau)J_{32}}}\rangle_{ss} &= \lim_{t\to\infty} \langle\overline{\overline{J_{23}(t+\tau)J_{32}(t)}}\rangle\,. \end{aligned} \tag{27}$$

We have assumed here that $\Omega \ll \omega_{31}$, so the Rayleigh-type and the Raman-type processes can be clearly separated. In fact the correlation functions (27) reflect this separation. The first one describes the Rayleigh-type scattering while the second one describes the Raman-type scattering (the Stokes line). According to the transformation (11), for the resonant case, we have

$$
\begin{aligned}
J_{21} &= \tfrac{1}{2}[R_{22} - R_{11} + R_{21} - R_{12}] , \\
J_{23} &= \frac{1}{\sqrt{2}} [R_{23} - R_{13}] ,
\end{aligned} \tag{28}
$$

which relates the "bare" atomic operators J_{21} and J_{23} to the "dressed" operators R_{ij}. Using the relations (28), the solutions of eqs. (22)–(25) for one-time averages, and applying the quantum regression theorem one obtains the following expressions for the correlation functions (27):

$$
\begin{aligned}
\langle \overline{\overline{J_{21}(\tau) J_{12}}} \rangle_{\mathrm{ss}} = \tfrac{1}{4}\{ &\langle (R_{22} - R_{11})^2 \rangle_{\mathrm{s}}\, \mathrm{e}^{-\Gamma_0 \tau} \\
&+ \langle R_{12} R_{21} \rangle_{\mathrm{s}}\, \mathrm{e}^{-2\mathrm{i}\Omega\tau - \Gamma_{12}(\tau)} + \langle R_{21} R_{12} \rangle_{\mathrm{s}}\, \mathrm{e}^{2\mathrm{i}\Omega\tau - \Gamma_{21}(\tau)} \} ,
\end{aligned} \tag{29}
$$

$$
\begin{aligned}
\langle \overline{\overline{J_{23}(\tau) J_{32}}} \rangle_{\mathrm{ss}} = \tfrac{1}{2}\{ &\langle R_{13} R_{31} \rangle_{\mathrm{s}}\, \mathrm{e}^{-\mathrm{i}(\Omega_3 + \Omega)\tau - \Gamma_{13}(\tau)} \\
&+ \langle R_{23} R_{32} \rangle_{\mathrm{s}}\, \mathrm{e}^{-\mathrm{i}(\Omega_3 - \Omega)\tau - \Gamma_{23}(\tau)} \} ,
\end{aligned} \tag{30}
$$

where

$$
\begin{aligned}
\Gamma_0 &= 2\gamma_{\mathrm{c}} + \gamma_{21} + \gamma_{23} + (\gamma_{23} - \gamma_{31})(N - \langle R \rangle_{\mathrm{s}}) , \\
\Gamma_{12}(\tau) &= \Gamma_{21}(\tau) \\
&= [\tfrac{3}{2}(\gamma_{\mathrm{c}} + \gamma_{21}) + \gamma_{23} + 4\epsilon^2/\gamma_{\mathrm{a}} + (\gamma_{23} - \gamma_{31})(N - \langle R \rangle_{\mathrm{s}})]\tau \\
&\quad + \frac{4\epsilon^2}{\gamma_{\mathrm{a}}^2} (1 - \mathrm{e}^{-\gamma_{\mathrm{a}}\tau}) , \\
\Gamma_{13}(\tau) &= \Gamma_{23}(\tau) \\
&= [\tfrac{1}{2}(\gamma_{\mathrm{c}} + \gamma_{21} + \gamma_{23}) + \gamma_{31} + \epsilon^2/\gamma_{\mathrm{a}} + \tfrac{1}{2}(\gamma_{23} - \gamma_{31})(N - 2\langle R \rangle_{\mathrm{s}})]\tau \\
&\quad + \frac{\epsilon^2}{\gamma_{\mathrm{a}}^2} (1 - \mathrm{e}^{-\gamma_{\mathrm{a}}\tau}) .
\end{aligned} \tag{31}
$$

The exact expressions for the weighting factors of the particular exponents are given in the appendix. The terms proportional to $(\gamma_{23} - \gamma_{31})$ in the width functions (31) are the collective terms and neglecting them we obtain the one-atom results that include, however, the effects of both the phase and amplitude fluctuations of the exciting laser field. Since the collective parts of the widths do not anyhow depend on the laser field fluctuations (at least for the resonant case and within the approximations used by us), the immediate result of our calculations is that the laser fluctuations affect only the one-atom parts of the widths. This is similar to the result obtained by Puri and Hassan[10]) for a system of two-level atoms. The correlation functions (29) and (30) have the well-known structure with the Mollow triplet for the Rayleigh line and the Autler–Townes splitting of the Stokes line. For the one-atom case, on neglecting the laser field fluctuations, our results agree with that of Cohen-Tannoudji and Reynaud[1]) and Agarwal and Jha[2]). For many atoms, however, the collective parts of the widths can become dominant. In fact, there are three qualitatively different cases when the number of atoms is large. From eq. (A.1) we have for $N \gg 1$

$$\langle R\rangle_s \rightarrow \begin{cases} N & \text{if } X > 1\,, \\ \frac{2}{3}N & \text{if } X = 1\,, \\ \dfrac{2X}{1-X} & \text{if } X < 1\,, \end{cases} \tag{32}$$

where $X = \gamma_{31}/\gamma_{23}$, and

$$(\gamma_{23} - \gamma_{31})(N - \langle R\rangle_s) \rightarrow \begin{cases} -\gamma_{23} & \text{if } X > 1\,, \\ 0 & \text{if } X = 1\,, \\ N(\gamma_{23} - \gamma_{31}) & \text{if } X < 1\,, \end{cases} \tag{33}$$

$$\tfrac{1}{2}(\gamma_{23} - \gamma_{31})(N - 2\langle R\rangle_s) \rightarrow \begin{cases} 0 & \text{if } X = 1\,, \\ \frac{1}{2}N|\gamma_{23} - \gamma_{31}| & \text{if } X \neq 1 \end{cases}\,. \tag{34}$$

So, for $X > 1$, i.e. $\gamma_{31} > \gamma_{23}$, practically all the population is shared by the atomic levels $|1\rangle$ and $|2\rangle$ and the collective narrowing of the one-atom Rayleigh lines takes place. In the limiting case the γ_{23} contribution to the one-atom width is completely canceled out by the collective contribution. In this case the weighting factors of the Rayleigh-type lines are proportional to N^2 and this part of the emitted radiation exhibits superradiant behaviour. The Stokes lines have the weighting factors proportional to N and their widths have broad ($\sim N$) collective components.

In the opposite case, $X < 1$, i.e. $\gamma_{31} < \gamma_{23}$, practically all the population is concentrated on the level $|3\rangle$. The weighting factors of the Rayleigh-type lines do not depend on N and only very weak radiation with a broad ($\sim N$) width is a remnant of the strongly driven $|1\rangle \rightarrow |2\rangle$ transition. The Stokes lines, as before, have the amplitudes and widths proportional to N.

In the particular case $X = 1$, the three atomic levels are equally populated, all the collective widths are zero, and all the weighting factors are proportional to N^2. Thus in this particular case also the Raman-type radiation exhibits superradiant behaviour.

One thus can say that, for $N \gg 1$, $X > 1$, the three-level atom system behaves like a two-level atom system when looking at the Rayleigh part of the emitted radiation. The effects of the phase and amplitude fluctuations of the laser field are exactly the same as for the two-level atom system[8,10]). The amplitude fluctuations affect only the sidebands while all the lines are affected by the phase fluctuations.

The Stokes lines are affected by both the phase and amplitude fluctuations of the exciting laser field.

5. Conclusions

We have considered the problem of collective resonance Raman scattering of an intense laser field from the point of view of the collective effects and the influence of the laser field fluctuations. The phase fluctuations of the field were modeled by the phase diffusion process while the amplitude fluctuations by a nonwhite Gaussian process. The master equation for the density matrix averaged over the phase and the amplitude fluctuations has been obtained in the secular approximation. It has been shown that for the exact resonance the master equation has a very simple solution. This solution has been used to calculate all needed steady-state averages. A decorrelation scheme has been used to obtain the solutions for the time-dependent averages, and the quantum regression theorem to deal with the two-time correlation functions. The field correlation functions have been calculated for the Rayleigh-type and the Raman-type processes. It has been shown that the laser field fluctuations affect only the one-atom parts of the spectral widths. The collective narrowing of the spectral lines has also been predicted. The collective (superradiant) properties of the emitted light have been discussed for various ranges of values of the atomic parameters. It was shown that only for $X = 1$ the Raman part of the emitted light exhibits superradiant behaviour.

Corresponding formulas for the intensity correlation functions can be obtained in a similar manner.

Appendix

In this appendix we give the explicit expressions for the steady-state averages of the atomic operators that have been calculated with the use of the density matrix (19). The normalization factor A appearing in the formulas is given by

$$A = [(N+1)X^{N+2} - (N+2)X^{N+1} + 1]/(X-1)^2 ,$$

where $X = \gamma_{31}/\gamma_{23}$, and the formulas are

$$\langle R \rangle_s = A^{-1}[N(N+1)X^{N+3} - 2(N+2)NX^{N+2} + (N+1)(N+2)X^{N+1} - 2X]/(X-1)^3 , \tag{A.1}$$

$$\langle R^2 \rangle_s = A^{-1}[N^2(N+1)X^{N+4} - N(3N^2+6N-1)X^{N+3} + (N+2)(3N^2+3N-2)X^{N+2} - (N+1)^2(N+2)X^{N+1} + 4X^2 + 2X]/(X-1)^4 , \tag{A.2}$$

$$\langle R_{22} \rangle_s = \langle R_{11} \rangle_s = \tfrac{1}{2}\langle R \rangle_s , \tag{A.3}$$

$$\langle R_{33} \rangle_s = N - \langle R \rangle_S , \tag{A.4}$$

$$\langle R_{33} - R_{22} \rangle_s = \langle R_{33} - R_{11} \rangle_s = N - \tfrac{3}{2}\langle R \rangle_s , \tag{A.5}$$

$$\langle R_{21}R_{12} \rangle_s = \langle R_{12}R_{21} \rangle_s = \tfrac{1}{2}\langle (R_{22} - R_{11})^2 \rangle_s = \tfrac{1}{6}[\langle R^2 \rangle_s + 2\langle R \rangle_s] , \tag{A.6}$$

$$\langle R_{13}R_{31} \rangle_s = \langle R_{23}R_{32} \rangle_s = \tfrac{1}{2}[(N+1)\langle R \rangle_s - \langle R^2 \rangle_s] . \tag{A.7}$$

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