

to a splitting  $\Delta\nu' = 695.0$  kHz, as against  $\Delta\nu'' = 698.2$  kHz.<sup>2</sup> This allows us to state that the experimentally observed value of  $\Delta\nu$  agrees with that calculated by us, since the difference between the splittings  $\Delta\nu'$  and  $\Delta\nu''$  lies within the limits of the measurement accuracy. Unfortunately, the splitting (19) turns out to be much less than the value that can be registered at present in experiment with two different rotating RR even if the Verdet constant is increased by two orders of magnitude compared with R of K-8 glass.

Thus, besides the good quantitative agreement of the theoretical analysis, proposed in the present paper, for the propagation of an electromagnetic field in rotating optically active media, with the experimental data, our experiment has confirmed the validity of relation (16), which is a statement of the additivity of the effect of non-reciprocity when account is taken of the rotation of a gyrotropic medium.

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## Polarization of resonance fluorescence of an atom with degenerate levels in a strong field

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The problem of resonance scattering of a monochromatic wave by an atom with allowance for saturation effects is solved in the radiation-field coherent-state representation. A general calculation scheme for scattering by a multilevel atom is presented. The exact solution for a two-level atom is identical with the results obtained by other authors. The method is applied to the problem of scattering of a linearly polarized light wave by two degenerate levels of the  $S_{1/2}$ - $P_{1/2}$  type in alkali metal atoms. The polarization characteristics of the process are discussed.

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### 1. INTRODUCTION

A number of papers are devoted to the question of the fluorescence spectrum of a two-level atom in the field of an intense monochromatic wave for times which are much larger than the lifetime of the free atom.<sup>1-15</sup>

There is a well-known expression for the probability of spontaneous emission of a photon with frequency  $\omega$  per unit frequency interval,<sup>3</sup> which in the case of high intensity of the incident field ( $\Omega \gg \gamma$ ,  $|\epsilon|$ ) takes the form

$$I_0(\omega) = \frac{\gamma/4}{(\omega - \omega_k)^2 + \gamma^2/4} + \frac{3\gamma/16}{(\omega - \omega_k - \Omega)^2 + 9\gamma^2/16} + \frac{3\gamma/16}{(\omega - \omega_k + \Omega)^2 + 9\gamma^2/16} \quad (1)$$

Here  $\omega_k$  is the frequency of the incident wave,  $\omega_0$  is the separation between levels,  $\gamma$  is the spontaneous width,  $\Omega = |E \cdot d|/\hbar$  is the Rabi frequency, and  $\epsilon = \omega_0 - \omega_k$

is the resonance detuning parameter.

We should point out that the results of work on resonance fluorescence substantially depend on the mathematical formulation and the physical set-up of the problem, and very often agree only when the exciting radiation is of high intensity. An analysis of the reasons for such a divergence of the results is presented in papers by Swain<sup>10</sup> and Raman.<sup>12</sup> One frequently used method for solving the problem is expansion of the wave function of the field in states with a fixed number of photons, and an approximate uncoupling of the infinite system of equations for the density matrix elements formed in this case.<sup>4-6,9-12</sup> We must approach the evaluation of the results obtained by this means very cautiously, since they depend substantially on the uncoupling

ling method employed.<sup>12</sup> A different approach<sup>2-3,14</sup> is based on factorization of the density matrix of the "atom + field" system (the Markov approximation). Such a procedure may be justified by the fact that changes occurring in the atom affect the vacuum field weakly.<sup>15</sup>

In this work we show that the use of dipole and resonance approximations allows us to carry out the solution of the problem of the resonance fluorescence spectrum to the end without drawing on any additional assumptions. The use of coherent field states plays an essential role in this presentation. The equations thus obtained for the matrix elements are analogous to the equations for an atom in a random external field whose amplitude has a Gaussian distribution.

## 2. COMPUTATIONAL TECHNIQUE

The radiation-field coherent-states representation is best suited for the mathematical formulation of the problem.<sup>16</sup> First of all, in this representation the equations for the matrix elements of the operators, as will be seen later, have a compact form. Secondly, the coherent states correspond better to the physical formulation of the problem. Indeed, if the radiation field is confined in a resonator, the number of photons in each mode is the convenient variable; in the situation under consideration, the radiation falls on each atom with a continuous flux and is scattered. Here, the potential of the field at the point at which the atom is located is an even more appropriate variable.

To start with, let us present the overall computational scheme for the case of a multilevel system—for example, a two-level system with an arbitrary degree of level degeneracy. The Hamiltonian of the system is written in the usual form ( $\hbar = c = 1$ )

$$H = \sum_{k\lambda} \omega_k C_{k\lambda} C_{k\lambda}^+ + \sum_m E_m a_m^+ a_m + \sum_{m,n} \sum_{k\lambda} (\beta_{m,n,k\lambda}^+ C_{k\lambda} + \beta_{m,n,k\lambda} C_{k\lambda}^+) a_m^+ a_n, \quad (2)$$

where conventional symbols are used:  $a_m^+$  and  $a_m$  are the creation and annihilation operators of the system in the  $m$ -th energy level;  $C_{k\lambda}^+$  and  $C_{k\lambda}$  are the photon creation and annihilation operators;  $\beta_{m,n,k\lambda}$  is the matrix element describing the coupling of the atom with the field.

The parameter which must be determined—the spectral density  $P(\omega)$ —is the number of photons scattered per unit time per unit frequency interval (i. e., a dimensionless parameter), and is expressed as follows:

$$P(\omega) = \frac{d}{dt} \sum_{k\lambda} \langle \alpha | \bar{C}_{k\lambda}^+(t) \bar{C}_{k\lambda}(t) | \alpha \rangle \delta(\omega_k - \omega), \quad (3)$$

where the vector  $|\alpha\rangle$  corresponds to the state of the atom and the radiation field as  $t \rightarrow -\infty$ ;  $\bar{C}_{k\lambda}^+$  and  $\bar{C}_{k\lambda}$  are the scattered field operators. On the other hand, using an arbitrary complete set of states  $|\beta\rangle$ , we may write

$$\begin{aligned} \langle \alpha | \bar{C}_{k\lambda}^+(t) \bar{C}_{k\lambda}(t) | \alpha \rangle &= \sum_{\beta} \langle \alpha | \bar{C}_{k\lambda}^+(t) | \beta \rangle \langle \beta | \bar{C}_{k\lambda}(t) | \alpha \rangle \rightarrow \\ &\rightarrow \sum_{\beta} \beta_{m,n,k\lambda}^+ \beta_{ij,k\lambda} \sum_{\gamma} \langle \alpha | \sigma_{m\gamma}^+(-\omega_k) | \beta \rangle \langle \beta | \sigma_{i\gamma}(\omega_k) | \alpha \rangle, \quad t \rightarrow \infty, \quad (4) \end{aligned}$$

where  $\sigma_{mn} = a_m^+ a_n$ .

In accordance with Eq. (4), in the equations of motion<sup>17</sup> for the operators  $\sigma_{mn}$  (they are not given here due to their unwieldiness) we go from the operators to their matrix elements between the initial  $|\alpha\rangle$  and intermediate  $|\beta\rangle$  states. We define the latter as follows:

$$|\alpha\rangle = \chi_{m_0} |f\rangle, \quad |\beta\rangle = \chi_{m_0} \prod_{k\lambda} |g_{k\lambda} + f \delta_{k\lambda} \delta_{k\lambda_0}\rangle, \quad (5)$$

where  $\chi_{m_0}$  is the wave function of the initial state of the atom ( $t \rightarrow -\infty$ ), and  $|f\rangle$  is the coherent state of the radiation field corresponding to the momentum  $k_0$  and the polarization  $\lambda_0$  (incident wave). In the intermediate states, it is convenient to separate out the initial amplitude  $f$ , then  $g_{k\lambda}$  represents the deviation from the initial values. Furthermore, we consider only those intermediate states in which the atom occurs in the state  $\chi_{m_0}$ , since the nondiagonal (relative to the atom) matrix elements of the operators satisfy a system of homogeneous equations and their contribution at  $t \rightarrow \infty$  vanishes.

We now make some simplifying assumptions, in order to eliminate unwieldy symbols. Let all the levels be connected by resonant transitions, which allows us to select oscillating exponentials for the matrix elements of the external field by a simple transformation. Then we will designate the matrix elements  $\langle \beta | \sigma_{mn} | \alpha \rangle$  as  $R_a$ , after excluding the corresponding exponentials. The  $R_a$  satisfy the following system of equations:

$$iR_a = \sum_b L_{ab} R_b - \sum_{k\lambda} \sum_{\beta} g_{k\lambda} B_{ab} R_b \exp[i(\omega_k - \omega_k) t] + Z \Gamma_a, \quad (6)$$

$$Z = \langle \beta | \alpha \rangle = \exp\left(-\frac{1}{2} \sum_{k\lambda} |g_{k\lambda}|^2 + i\varphi\right). \quad (7)$$

Here the time-independent matrix  $L_{ab}$  describes the interaction with the external field and the relaxation processes, and the matrix  $B_{ab}$  (also time-independent) is composed of the matrix elements  $\beta_{m,n,k\lambda}$ ; we may drop the insignificant phase  $\varphi = 2 \text{Im}(f g_{k_0 \lambda_0})$ .

From Eq. (6) we obtain for the Fourier components of  $R_a$

$$\begin{aligned} R_a(\omega - \omega_k) &= - \sum_{k\lambda} \sum_{\beta} g_{k\lambda} (\omega - \omega_k - \mathcal{L})_{ab}^{-1} B_{bc} R_c(\omega + \omega_k - 2\omega_k) \\ &\quad - Z \sum_{\beta} (L)_{ab}^{-1} \Gamma_b \delta(\omega - \omega_k). \end{aligned} \quad (8)$$

Eq. (4) may be rewritten now as follows:

$$\begin{aligned} \langle \alpha | \bar{C}_{k\lambda}^+(t) \bar{C}_{k\lambda}(t) | \alpha \rangle &\rightarrow \\ &\rightarrow \sum_{\beta} \beta_{m,n,k\lambda}^+ \beta_{ij,k\lambda} \int R_a^*(\omega_k - \omega_k) R_b(\omega_k - \omega_k) \prod_{k'\lambda'} d^2 g_{k'\lambda'}, \quad t \rightarrow \infty. \quad (9) \end{aligned}$$

If we set all  $g_{k\lambda} = 0$  in Eq. (6), then, as was to be expected, we obtain the usual equations presented by Milonni and Smith<sup>17</sup> for the density matrix. The parameter  $g_{k\lambda}$  appears in the role of the amplitude of an unusual noise field with a Gaussian distribution. Indeed, the inhomogeneous term in system (6) contains  $\exp(-\frac{1}{2} (\sum |g_{k\lambda}|^2))$ , meaning that

$$R_a^*(\omega) R_b(\omega) \sim \exp(-\sum |g_{k\lambda}|^2)$$

and the summation over the intermediate states  $|\beta\rangle$ , which according to Eq. (9) is reduced to integration over

$|g_{\mathbf{k}\lambda}|$  and  $\arg g_{\mathbf{k}\lambda}$ , means averaging over the Gaussian distribution. The noise field is unusual because it enters into the system in Eq. (6) in a "non-Hermitian" manner; i.e., only its positive-frequency part is involved. Thus, use of coherent states allows us to reduce the problem to the system (8), eliminating the difficulties associated with the quantum character of the field.

Using the obvious relations  $\langle Z^2 g_{\mathbf{k}\lambda} \rangle = 0$  and  $\langle Z^2 g_{\mathbf{k}\lambda}^* g_{\mathbf{k}\lambda} \rangle = \delta_{\mathbf{k}\mathbf{k}'} \delta_{\lambda\lambda'}$ , where  $\langle \dots \rangle$  indicates averaging of Eq. (9) over the states of the intermediate field  $g_{\mathbf{k}\lambda}$ , we may show by means of a power series expansion of  $g_{\mathbf{k}\lambda}$  and term-by-term averaging the validity of the following relationship:

$$\langle g_{\mathbf{k}\lambda}^* g_{\mathbf{k}\lambda} R_a^*(\nu + \omega_{\mathbf{k}}) R_b(\nu + \omega_{\mathbf{k}}) \rangle = \delta_{\mathbf{k}\mathbf{k}'} \delta_{\lambda\lambda'} \langle R_a^*(\nu + \omega_{\mathbf{k}}) R_b(\nu + \omega_{\mathbf{k}}) \rangle, \quad (10)$$

corresponding to lack of retardation effects and leading to loss of memory.<sup>3</sup> In retaining this expression, the weak dependence of the matrix element  $\beta_{m n, \mathbf{k}\lambda}$  on frequency was neglected (the dipole approximation) and the quantization volume  $v$  tended toward infinity (recall that  $\beta_{m n, \mathbf{k}\lambda} \sim v^{-1/2}$ ), which also led to vanishing of the retardation effects. We note, however, that the approximation involving a weak frequency dependence for  $\beta_{m n, \mathbf{k}\lambda}$  has already been used from the beginning to obtain the equation of motion<sup>17</sup> for the atomic operators  $\sigma_{m n}$ , so that repetition of its use does not introduce additional approximations into the problem.

Bearing in mind Eq. (10) and taking into account the resonance character of the interaction, we may now write:

$$\langle R_a^*(\omega - \omega_{\mathbf{k}}) R_b(\omega - \omega_{\mathbf{k}}) \rangle = \sum_{bb'} (\hat{L}')_{ab}^{-1} (\hat{L}')_{a'b'}^{-1} \Gamma_b \Gamma_{b'} \delta(0) \delta(\omega - \omega_{\mathbf{k}}) + \sum_{bb'cc'} (\omega - \omega_{\mathbf{k}} - \hat{L}')_{ab}^{-1} (\omega - \omega_{\mathbf{k}} - \hat{L}')_{a'b'}^{-1} B_{bc} B_{b'c'} Q_{cc'}, \quad (11)$$

where  $\hat{Q}$  satisfies the equation

$$Q_{aa'} = \sum_{bb'} (\hat{L}')_{ab}^{-1} (\hat{L}')_{a'b'}^{-1} \Gamma_b \Gamma_{b'} \delta(0) + \sum_{bb'cc'} \left[ \int_{-\infty}^{\infty} (x - \hat{L}')_{ab}^{-1} (x - \hat{L}')_{a'b'}^{-1} dx \right] B_{bc} B_{b'c'} Q_{cc'}. \quad (12)$$

Thus, the problem of determining the resonance fluorescence spectrum in a strong field is reduced to the inversion of the matrix  $x - \hat{L}$  and the solution of the matrix equation for  $\hat{Q}$  [Eq. (12)]. In a particular case there follows from (11) and (12), for the fluorescence spectrum of a nondegenerate two-level system, an expression that agrees exactly with the results of Mollow<sup>3</sup>; and in the case  $\varepsilon^2 + \Omega^2 \gg \gamma^2$ , also with the results of Ref. 13. The limiting form of this expression for  $\Omega \gg \gamma$ ,  $|\varepsilon|$  is given by Eq. (1).

### 3. ALLOWANCE FOR LEVEL DEGENERACY

Let us now turn to the study of the fluorescence spectrum of a two-level system  $S_{1/2} - P_{1/2}$  placed in the field of a linearly polarized monochromatic wave, taking into account the level degeneracy. We note immediately that the presence of degeneracy in such a system does not lead to an increase in the number of peaks in the fluorescence spectrum; since only transitions with  $\Delta m = 0$  are induced by the wave field (the

quantization axis is chosen along the direction of the polarization vector of the incident wave), whereas mixing of degenerate sublevels occurs only as a result of spontaneous transitions with  $\Delta m = \pm 1$ , i.e., additional Rabi frequencies do not arise. Nevertheless, it is quite essential to take the degeneracy into account, since its existence, as will be seen later on, leads to the development of an angular dependence, and to a change in amplitudes and widths of the peaks of the fluorescence spectrum.

From Eqs. (11) and (12), we obtain for the spectral density of photons scattered per unit time with momentum  $\mathbf{k}$  and polarization  $\lambda$ :

$$P(\mathbf{k}, \lambda) = \frac{\gamma}{2\pi} I_0(\omega_{\mathbf{k}}) |\mathbf{e}_0 \mathbf{e}_{\mathbf{k}\lambda}|^2 + \frac{\gamma}{2\pi} I_1(\omega_{\mathbf{k}}) (1 - |\mathbf{e}_0 \mathbf{e}_{\mathbf{k}\lambda}|^2). \quad (13)$$

Here  $\mathbf{e}_0$  and  $\mathbf{e}_{\mathbf{k}\lambda}$  are the polarization vectors of the incident and scattered fields; the spectral function  $I_0(\omega)$  in the case  $\Omega \gg \gamma$ ,  $|\varepsilon|$  is described by Eq. (1) (its exact form in the case of an arbitrary relationship of the parameters  $\Omega$ ,  $\varepsilon$ , and  $\gamma$  is given in Ref. 3) and the function  $I_1(\omega)$  has the form

$$I_1(\omega) = \frac{2}{3} \gamma \Omega^2 n \frac{A + B(\omega - \omega_{\mathbf{k}})^2 + (\omega - \omega_{\mathbf{k}})^4}{|F(i(\omega - \omega_{\mathbf{k}}))|^2}, \quad (14)$$

where

$$n = \frac{1}{4} \Omega^2 \left/ \left( \varepsilon^2 + \Omega^2 + \frac{1}{4} \gamma^2 \right), \right. \\ F(x) = x^4 + 2\gamma x^3 + \left( \varepsilon^2 + \Omega^2 + \frac{5}{4} \gamma^2 \right) x^2 + \gamma \left( \varepsilon^2 + \frac{5}{6} \Omega^2 + \frac{1}{4} \gamma^2 \right) x + \frac{1}{6} \gamma^2 \Omega^2, \quad (15)$$

and the constants  $A$  and  $B$  are determined from the relationship

$$\frac{1}{2\pi} \int_0^{\infty} I_1(\omega) d\omega = n, \quad \frac{1}{2\pi} \int_0^{\infty} I_1(\omega) (\omega - \omega_{\mathbf{k}})^2 d\omega = \frac{1}{2} n \Omega^2. \quad (16)$$

The first term in Eq. (13) describes the radiation scattered in transitions between atomic levels which are connected by the external field ( $\Delta m = 0$ ). The component  $I_1(\omega)$  of this expression describes the radiation scattered in transitions with  $\Delta m = \pm 1$ .

The polarization characteristics of the scattered radiation, determined by Eq. (13), in certain particular cases have rather simple characteristics, which allow us to separate out the spectral components  $I_0(\omega)$  and  $I_1(\omega)$  in the observation. Thus, for example, in the  $z$  direction (the  $z$  axis is chosen in the direction of the polarization vector of the incident wave  $\mathbf{e}_0$ , the  $x$  and  $y$  axes in this case are chosen arbitrarily) we will observe naturally polarized light with the  $I_1(\omega)$  spectrum. The scattered radiation propagating along the  $x$  axis consists of two components: a linearly polarized component along the  $z$  axis with spectrum  $I_0(\omega)$  and a linearly polarized component along the  $y$  axis with spectrum  $I_1(\omega)$ . Analysis of the polarization properties of the scattered radiation, but for  $t \ll \gamma^{-1}$ , was carried out by Adonts and Kocharyan.<sup>18</sup> For longer observation times ( $t \gg \gamma^{-1}$ ), an analogous analysis (but only for the case  $\varepsilon^2 + \Omega^2 \gg \gamma^2$ ) is given in Ref. 19.

Summing over the polarizations in Eq. (13), we obtain an expression for the angular distribution of the

scattered radiation:

$$dP(\omega) = \gamma [I_0(\omega) \sin^2 \theta + I_1(\omega) (1 + \cos^2 \theta)] d\theta / 4\pi, \quad (17)$$

where  $\theta$  is the angle between the  $z$  axis and the direction of observation.

The behavior of the function  $I_0(\omega)$  in various limiting cases has been rather thoroughly investigated by Mollow<sup>3</sup>; therefore we have decided to only analyze the limiting forms of the spectral component  $I_1(\omega)$ . In the case of low intensity of the exciting radiation and a large detuning parameter ( $|\varepsilon| \gg \gamma \gg \Omega$ ), Eq. (14) is transformed into

$$I_1(\omega) = \frac{\Omega^2}{4\varepsilon^2} \left\{ 2\pi \delta(\omega - \omega_k) + \frac{\gamma \Omega^2}{4\varepsilon^2} \left[ \frac{1}{(\omega - \omega_k - \varepsilon)^2 + \gamma^2/4} + \frac{1}{(\omega - \omega_k + \varepsilon)^2 + \gamma^2/4} \right] \right\}. \quad (18)$$

The primary contribution to the integrated intensity in this case comes from the scattering at the unshifted frequency.

In the other limiting case of high intensity of the incident field ( $\Omega \gg \gamma, |\varepsilon|$ ), Eq. (14) takes on the form

$$I_1(\omega) = \frac{\gamma/4}{(\omega - \omega_k)^2 + \gamma^2/4} + \frac{7\gamma/48}{(\omega - \omega_k - \Omega)^2 + (7\gamma/12)^2} + \frac{7\gamma/48}{(\omega - \omega_k + \Omega)^2 + (7\gamma/12)^2}. \quad (19)$$

It is interesting to note that the central peak has a Lorentzian shape only for this case. It is interesting to compare the heights of the central peak (1) and of the satellites (3/7), as well as their integrated intensities ( $\frac{1}{2}\gamma$  and  $\frac{1}{4}\gamma$ ) of the  $I_1(\omega)$  component with the corresponding values 1, 1/3 and  $\frac{1}{2}\gamma, \frac{1}{4}\gamma$  for  $I_0(\omega)$ .

The problem of determining the fluorescence spectrum is substantially simplified when the Rabi-nutation frequency for any allowed atomic transition is significantly greater than its spontaneous width (for the two-level system, this condition has the form  $\varepsilon^2 + \Omega^2 \gg \gamma^2$ ). This condition, indicating nonoverlapping peaks in the fluorescence spectrum, is satisfied in particular when the intensity of the external field reaches its maximum

value, i.e., when the magnitude of the dynamic Stark shift of any level is much greater than its spontaneous width. For this case, a method has been proposed,<sup>13</sup> which allows comparatively simple calculation of the integrated intensities of the scattered field lines. The widths of these lines, as follows from Eqs. (11) and (12), may be easily determined by calculating the imaginary components of the roots of the determinant of the  $x - \hat{L}$  matrix.

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