Spectral properties of the Hanle effect for a strong monochromatic wave

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Spectral features of the Hanle effect are investigated for the monochromatically excited atomic system $\mathbf{J}=0\rightarrow\mathbf{J}=1$. Explicit expressions for the spectrum of the Hanle signal are found. It is shown that the Hanle signal spectrum like the fluorescence spectrum of a two-level atom contains both elastic and inelastic components. There may be up to seven peaks in the spectrum of the inelastic component. An analysis of the fluorescence spectra is presented in terms of dressed-atom states.

1. INTRODUCTION

A knowledge of features of the Hanle effect[1,2] is of great importance in the laser spectroscopy of multilevel atomic systems since it is an analog of transitions between excited states which are becoming mutually coherent. A simple, albeit very useful, system relevant to the interaction of radiation with atoms is the two-level atom.[3] In the investigation of the Hanle effect, the transition $\mathbf{J}=0\rightarrow\mathbf{J}=1$ plays such a role and is considered in the present paper.

The typical experimental geometry for the Hanle effect is shown in Fig. 1: the incident wave is linearly polarized along the observation axis of the fluorescence signal. In zero magnetic field the signal is absent since the radiating transitions from the $m_J=\pm 1$ sublevels cancel each other out as a result of interference. In a magnetic field these transitions begin to evolve with different frequencies, precession of the polarization appears, and fluorescence is observed.

The Hanle effect was observed in a monochromatically excited atom by Rasmussen, Schieder, and Walther[4] at an intensity small in comparison with the saturation intensity. The theoretical analysis of Avan and Cohen-Tannoudji[5] applies to a wave of arbitrary intensity; even so, they did not investigate the spectral aspects of the fluorescence. This was not even done in Ref. 6, where the thermal motion of the atoms was assumed to be a decisive factor and, as in Ref. 5, attention was given only to the total intensity of the scattered radiation.

The resonance fluorescence spectrum for a monochromatic wave of arbitrary intensity was first obtained by Rautian and Sobel'man[7] for an open two-level system and by Mollow[8] for a two-level atom. To describe the Hanle effect, we have used the results of Refs. 9–11, which are based on the atom-photon density matrix formalism of Scully and Lamb.[12]

2. BASIC EQUATIONS

In the present section we present the basic equations which describe the interaction of resonance radiation with atoms subject to a constant magnetic field and laser radiation (see Fig. 1). In the rotating wave approximation and the dipole coupling approximation the Hamiltonian of the system under consideration has the form (in rad/s)

$$H=\Delta_L R_e+\sum_{j,\sigma}\nu_j a_{j\sigma}^{+}a_{j\sigma}+\Omega R_z+\sum_{\sigma}(V_\sigma R_{\sigma 0}^{+}+V_\sigma^{*}R_{0\sigma}^{-})$$
$$+\sum_{j,\sigma} i(g_{j\sigma}R_{\sigma 0}^{+}U_{j\sigma}a_{j\sigma}-g_{j\sigma}^{*}R_{0\sigma}^{-}U_{j\sigma}^{*}a_{j\sigma}^{+}), \qquad (1)$$

where $a_{j\sigma}$ is the annihilation operator for photons in the jth mode with wave vector $\mathbf{k}_{j\sigma}$ with polarization σ (left is "+" and right is "−") and frequency ω_j. In these expressions $\Delta_L=\omega_0-\omega_L$, $\nu_j=\omega_j-\omega_L$, ω_0 is the transition frequency, and ω_L is the frequency of the laser wave, which is linearly polarized parallel to the y axis (see Fig. 1). The third term in Eq. (1) describes the action of the magnetic field on the atoms; here Ω is the Larmor frequency. The fourth term describes the interaction of the atoms with the laser wave, which we will represent in the form of a sum of waves of left and right polarization with electric field intensity E_σ. Here we have put $V_\sigma=-\mu_{\sigma 0}E_\sigma U_L/2\hbar$, where $\mu_{\sigma 0}$ is the matrix element of the dipole moment at the transition $|0\rangle\rightarrow|\sigma\rangle$. Here the vector $|0\rangle$ describes the lower state of the atom, and $|\sigma\rangle$, the upper state, which is $|+\rangle$ for

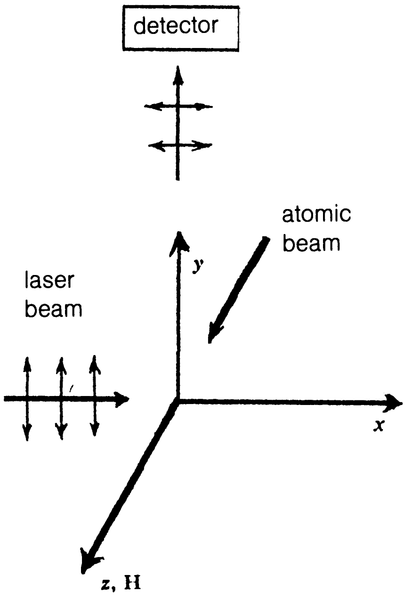

FIG. 1. Typical experimental setup for observing the Hanle effect.

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$m_J=\pm 1$ and $|-\rangle$ for $m_J=-1$. In the calculations we assume $|V_+|=|V_-|=V$. The quantity $U_j=U_j(\mathbf{r})$ describes the spatial mode factor;

$$g_{j\sigma}=\mu_{0\sigma}(2\pi\omega_j/c\hbar v)^{1/2}$$

is the coupling constant, and v is the quantization volume. In the description of the atomic system we make use of the matrix R, which has the form

$$R_e=\frac{1}{2}\begin{pmatrix}1&0&0\\0&1&0\\0&0&-1\end{pmatrix},\quad R_z=\begin{pmatrix}1&0&0\\0&-1&0\\0&0&0\end{pmatrix},$$
$$R^+_{+0}=\begin{pmatrix}0&0&1\\0&0&0\\0&0&0\end{pmatrix},\quad R^+_{-0}=\begin{pmatrix}0&0&0\\0&0&1\\0&0&0\end{pmatrix},\qquad (2)$$
$$R^-_{0+}=\begin{pmatrix}0&0&0\\0&0&0\\1&0&0\end{pmatrix},\quad R^-_{0-}=\begin{pmatrix}0&0&0\\0&0&0\\0&1&0\end{pmatrix}.$$

The components of the atomic density matrix ρ are defined as follows:

$$\rho_{\sigma 0}=\langle R^+_{\sigma 0}\rho\rangle,\quad \rho_{\sigma\sigma}=\langle R^+_{\sigma 0}R^-_{0\sigma}\rho\rangle,$$
$$\rho_{+-}=\langle R^+_{+0}R^-_{0-}\rho\rangle,\quad \rho_{00}=\langle R^-_{0\sigma}R^+_{\sigma 0}\rho\rangle. \qquad (3)$$

We define the atom–photon operator ρ_{a-f} of the atom + field system, for which we have the standard equation of motion:[10–12]

$$i\dot\rho_{a-f}=[H;\rho_{a-f}]+i\Gamma(\rho_{a-f}), \qquad (4)$$

where the operator $\Gamma(\rho_{a-f})$ describes relaxation processes. For the atomic density matrix $\boldsymbol{\rho}$, disregarding the interaction with the quantized modes, we have the following equation of motion:

$$i\dot{\boldsymbol{\rho}}=\mathbf{M}\boldsymbol{\rho}, \qquad (5)$$

where $\boldsymbol{\rho}^T=(\rho_{++},\rho_{--},\rho_{00},\rho_{0+},\rho_{+0},\rho_{0-},\rho_{-0},\rho_{+-},\rho_{-+})$, and the matrix $\mathbf{M}$ has the form

$$\mathbf{M}=\begin{vmatrix}
-i\gamma & 0 & 0 & V_+^* & -V_+ & 0 & 0 & 0 & 0\\
0 & -i\gamma & 0 & 0 & 0 & V_-^* & -V_- & 0 & 0\\
i\gamma & i\gamma & 0 & -V_{+-}^* & V_+ & -V_-^* & V_- & 0 & 0\\
V_+ & 0 & -V_+ & \Delta_+^* & 0 & 0 & 0 & 0 & V_-\\
-V_+^* & 0 & V_+^* & 0 & -\Delta_+ & 0 & 0 & -V_-^* & 0\\
0 & V_- & -V_- & 0 & 0 & \Delta_-^* & 0 & V_+ & 0\\
0 & -V_-^* & V_-^* & 0 & 0 & 0 & -\Delta_- & 0 & -V_+^*\\
0 & 0 & 0 & 0 & -V_- & V_+^* & 0 & \delta^* & 0\\
0 & 0 & 0 & V_-^* & 0 & 0 & -V_+ & 0 & -\delta
\end{vmatrix}, \qquad (6)$$

where $\Delta_\pm=\omega_L-\omega_0\mp\Omega+i\gamma/2$, $\delta=2\Omega+i\gamma$, and γ is the spontaneous relaxation rate. Since the atomic subsystem is closed, the total probability is conserved, and the system (5), which is linearly dependent, must satisfy the condition $\mathrm{Tr}(\boldsymbol{\rho})=1$.

The photon field operator P is defined on the basis of the operator ρ_{a-f} by taking the trace over the atomic states. The equation of motion for the slowly varying component of this operator to second order in the perturbation theory expansion over the coupling constant g has the form[9–11]

$$\dot P=\sum_{j,\sigma,\sigma'}[A_{j\sigma,j\sigma'}(a^+_{j\sigma}Pa_{j\sigma'}-Pa_{j\sigma'}a^+_{j\sigma})$$
$$+B_{j\sigma,j\sigma'}(a_{j\sigma'}Pa^+_{j\sigma}-a^+_{j\sigma}a_{j\sigma'}P)]. \qquad (7)$$

In this equation the terms which describe four-wave mixing (see Ref. 9) are absent. This is because the geometry of the Hanle effect is such (see Fig. 1) that the conditions for wave synchronism in the four-wave mixing of the scattered photons with the incident wave are absent.

In the problem under consideration, along with the average $\langle a^+_{j\sigma}a_{j\sigma}\rangle=\langle a^+_{j\sigma}a_{j\sigma}P\rangle=n_{j\sigma}$ (the occupation number of the photons with wave vector $\mathbf{k}_{j\sigma}$), the correlators $\langle a^+_{j\sigma'},a_{j\sigma}\rangle$ $(\sigma\neq\sigma')$ are also important. As was noted in Ref. 9, for these correlators the wave synchronism condition need not hold.

The equations of motion for the photon occupation numbers and the quantum correlators of photons of different polarization are

$$\frac{d}{dt}n_\sigma=\alpha_\sigma n_\sigma+\beta_\sigma\langle a^+_{\sigma'}a_\sigma\rangle+A_{\sigma\sigma}+\text{h.c.}, \qquad (8)$$

$$\frac{d}{dt}\langle a^+_{\sigma'}a_\sigma\rangle=(\alpha_{\sigma'}+\alpha^*_\sigma)\langle a^+_{\sigma'}a_\sigma\rangle+\beta_{\sigma'}n_\sigma+\beta^*_\sigma n_{\sigma'}$$
$$+A_{\sigma'\sigma}+A^*_{\sigma\sigma'}, \qquad (9)$$

where $\alpha_\sigma=A_{\sigma\sigma}-B_{\sigma\sigma}$ and $\beta_\sigma=A_{\sigma\sigma'}-B_{\sigma\sigma'}$ and it is as-

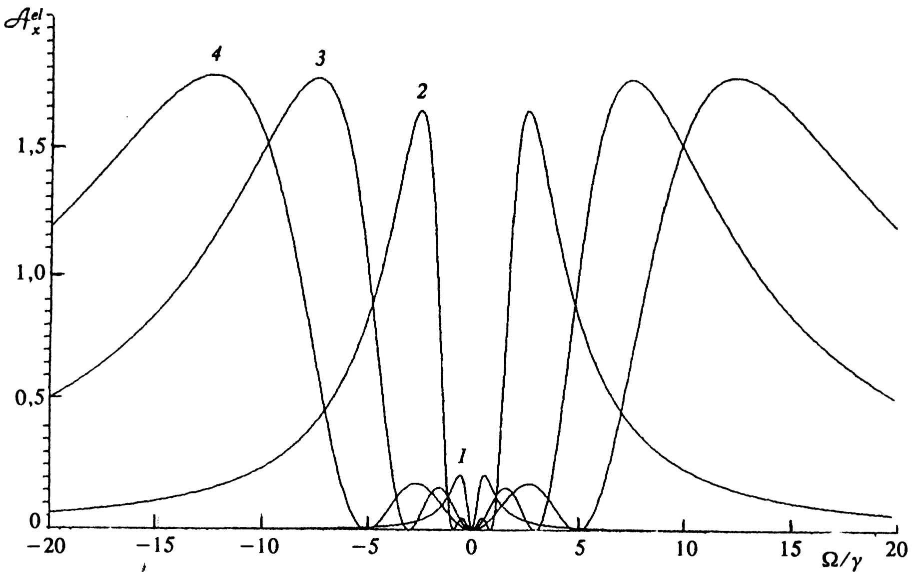

FIG. 2. Dependence of the elastic component of the Hanle signal on the magnetic field for $\Delta_L=0$ and $V/\gamma=0.1$ (*1*), 1 (*2*), 3 (*3*), 5 (*4*).

sumed that $\sigma\neq\sigma'$. Here and in what follows the index j is dropped since, as it turns out, the spectrally degenerate modes are coupled.

The coefficients A and B are defined as follows:

$$A_{++}=iN|g_+|^2(z_{43}\rho_{+0}-z_{44}\rho_{++}-z_{46}\rho_{+-}), \tag{10}$$

$$B_{++}=iN|g_+|^2(-z_{44}\rho_{00}+z_{41}\rho_{+0}+z_{49}\rho_{-+}), \tag{11}$$

$$A_{+-}=iNg_-g_+^*(z_{43}\rho_{-0}-z_{44}\rho_{-+}-z_{46}\rho_{--}), \tag{12}$$

$$B_{+-}=-iNg_-g_+^*(z_{42}\rho_{-0}+z_{46}\rho_{00}+z_{48}\rho_{+-}), \tag{13}$$

where ρ_{jk} is the stationary solution of system (5), $z_{jk}=\det(Z_{jk})/\det(\mathbf{Z})$, Z_{jk} is the jkth minor of the matrix $\mathbf{Z}=\mathbf{M}+\nu\mathbf{I}$ (here $\mathbf{I}$ is the unit matrix), $\nu=\omega-\omega_L$, and N is the number of atoms. The corresponding coefficients for $\sigma=$"$-$" or σ and σ' reversed are obtained by making the replacements $\Omega\rightarrow-\Omega$ and $V_+\leftrightarrow V_-$.

3. DESCRIPTION OF THE HANLE EFFECT

The proposed theory allows one to describe the Hanle effect, i.e., fluorescence in the system shown in Fig. 1. We have obtained equations for the occupation numbers of the photons of left and right polarization n_+ and n_-, the fluorescence spectra of which are determined by the quantities $\mathscr{A}_{++}$ and $\mathscr{A}_{--}$, where $\mathscr{A}_{\sigma\sigma'}=A_{\sigma\sigma'}+A^*_{\sigma'\sigma}$. Here the Hanle signal is determined by the occupation numbers of the photons of linear x-polarization, and the fluorescence spectrum, is determined by $\mathscr{A}_x$. Making use of the relations between the creation operators of circularly and linearly polarized photons, we have the following relations between the photon occupation numbers:[13]

$$n_x=n_++n-\langle a_+^+a_-\rangle-\langle a_-^+a_+\rangle, \tag{14}$$

$$n_y=n_++n_-+\langle a_+^+a_-\rangle+\langle a_-^+a_+\rangle. \tag{15}$$

The x-polarized photons are observed in the y direction, and the y-polarized photons, in the z direction (see Fig. 1). Thus, for an optically thin medium

$$n_x\propto\mathscr{A}_x=\mathscr{A}_{++}+\mathscr{A}_{--}-\mathscr{A}_{+-}-\mathscr{A}_{-+}, \tag{16}$$

$$n_y\propto\mathscr{A}_y=\mathscr{A}_{++}+\mathscr{A}_{--}+\mathscr{A}_{+-}+\mathscr{A}_{-+}. \tag{17}$$

The total intensity of the Hanle signal is $J_x\propto\int n_x d\nu$ or $J_x\propto\int\mathscr{A}_x d\nu$, respectively.

From the expressions for the spontaneous sources one can easily obtain the relation

$$\int\mathscr{A}_{\sigma\sigma'}d\nu\propto\rho_{\sigma\sigma'}, \tag{18}$$

which has a simple physical explanation: the fluorescence is proportional to the population of the excited level. Thus, for the intensity of the fluorescence signal we have the relations

$$J_x\propto\rho_{++}+\rho_{--}-\rho_{+-}-\rho_{-+}, \tag{19}$$

$$J_y\propto\rho_{++}+\rho_{--}+\rho_{+-}+\rho_{-+}. \tag{20}$$

This result was obtained earlier by Avan and Cohen-Tannoudji.[5]

As is known,[8] the resonance fluorescence spectrum of a two-level atom contains an elastic, or unshifted, component which is proportional to $\delta(\nu)$, and an inelastic component which describes scattering with change of the frequency of the quantum. This picture is also valid in the present case:

$$\mathscr{A}_{\sigma\sigma'}=\mathscr{A}^{\mathrm{el}}_{\sigma\sigma'}+\mathscr{A}^{\mathrm{inel}}_{\sigma\sigma'}, \tag{21}$$

$$\mathscr{A}_x=\mathscr{A}^{\mathrm{el}}_x+\mathscr{A}^{\mathrm{inel}}_x. \tag{22}$$

The elastic component $\mathscr{A}^{\mathrm{el}}_{\sigma\sigma'}$ is given by the expression

$$\mathscr{A}^{\mathrm{el}}_{\sigma\sigma'}=2\pi Ng_{\sigma'}g^*_{\sigma}\rho_{0\sigma}\rho_{\sigma'0}\delta(\nu). \tag{23}$$

The elastic component of the Hanle signal $\mathscr{A}^{\mathrm{el}}_x$, consequently, is given by the expression

$$\mathscr{A}^{\mathrm{el}}_x=2\pi N|g_+\rho_{+0}-g_-\rho_{-0}|^2\delta(\nu). \tag{24}$$

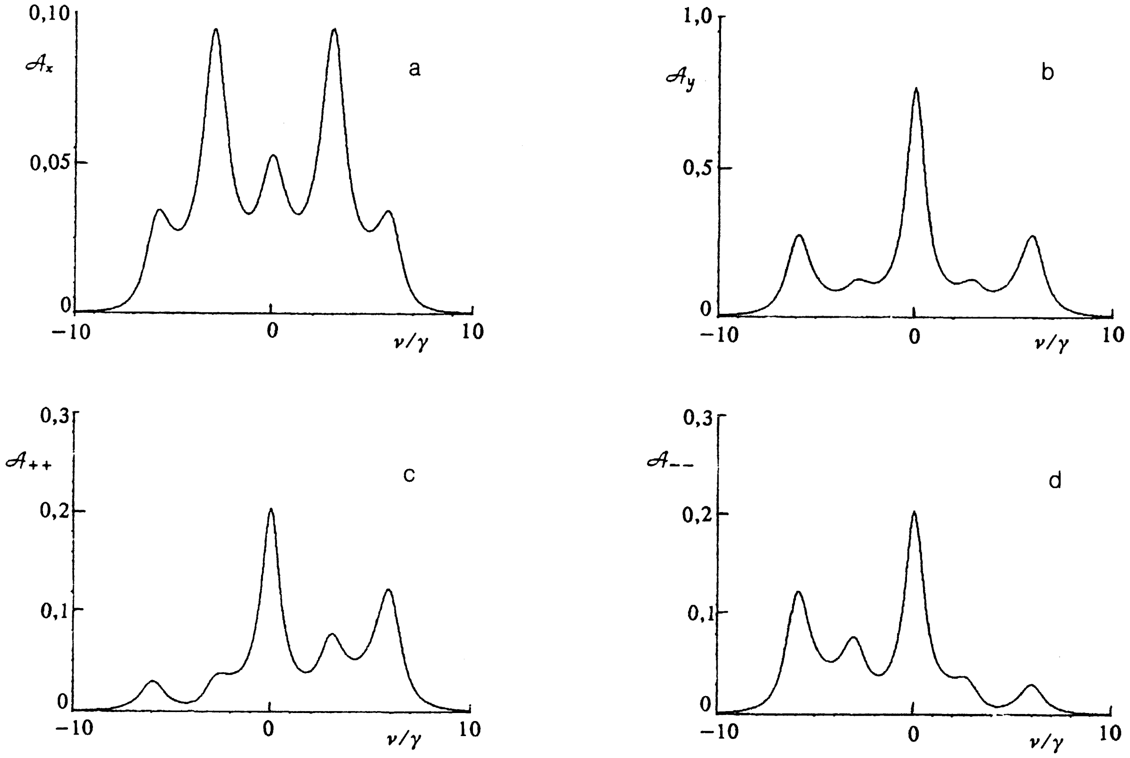

FIG. 3. Spectral dependences of spontaneous sources for photons with a) x-polarization, b) y-polarization, c) left polarization, and d) right polarization, for $\Delta_L=0$, $V/\gamma=2$, and $\Omega/\gamma=1$.

4. FLUORESCENCE SPECTRA

In weak electromagnetic fields $V\ll\gamma$, the unshifted component dominates in the Hanle signal, just as in the spectrum of a two-level atom.[8] It is for this reason that the dependence of the intensity of the elastic component on the magnitude of the magnetic field shown in Fig. 2 (curve *1*) agrees with the dependence of the total scattering intensity in the absence of saturation derived in Ref. 5. In very strong magnetic fields $(\Omega/V\gg1)$ the elastic component can predominate even for saturating electromagnetic fields. This is because the magnetic field spreads the levels so far apart that the excitation becomes weak and sufficiently strong mixing of the excited and ground states does not take place, as a result of which a shifted component appears in the scattering spectrum, and in strong fields the spectrum has a multipeak structure. In general, the ratio $V/|\Delta_\pm|$ may serve as a parameter of the separation of the spectrum into an elastic and inelastic component: for $V/|\Delta_\pm|\ll1$ the first of these dominates, and for $V/|\Delta_\pm|\gg1$, the second. Note that in saturating fields, such that $\Omega\approx\sqrt{2}V$ holds, the inelastic component vanishes (see Fig. 2). It may be supposed that this is a manifestation of the intersection of quasi-energy levels: as is well known, in an intense electric field the atomic levels split and undergo a shift—the high-frequency Stark effect. In the problem considered here, for $\Omega\approx\sqrt{2}V$ the level shifts due to the Zeeman effect in the one case and to the high-frequency Stark effect in the other turn out to be the same and, apparently, intersection of levels is observed for the inelastic component,[2] albeit under unusual conditions. This effect also takes place when the laser radiation is detuned from resonance $(\Delta_L\neq0)$, but for a different ratio between Ω and V.

Figure 3 shows the inelastic components of the polarization spectra for both linearly and circularly polarized photons under identical conditions. The spectrum of the photons with y-polarization (the same polarization as that of the pumping wave) is close to the Mollow spectrum since the magnitude of the magnetic field is relatively small. For this reason, the Hanle signal is small in comparison with the spectra of the other polarizations. The variation of the spectrum of the Hanle signal as a function of the magnitude of the magnetic field is shown in Figs. 4 and 5. From Fig. 4 it can be seen that the Hanle signal is small for both weak $(\Omega<\gamma)$ and strong fields $(\Omega>\gamma)$. In the first case this is because the levels are spread apart only slightly and emission is strongly suppressed by interference. In the second case, conversely, the levels are too greatly spread apart and the degree of excitation of the working transitions is diminished. It can be seen from Fig. 5 that in the intermediate region $\Omega\sim V>\gamma$ the Hanle signal reaches its greatest values.

It is well known that the resonance fluorescence spectrum can be interpreted as transitions between quasi-energy levels which are formed as a result of the action of an electromagnetic wave on the atomic system (states of the dressed atom). The quasi-energy spectrum in the system under consideration can be easily obtained from ex-

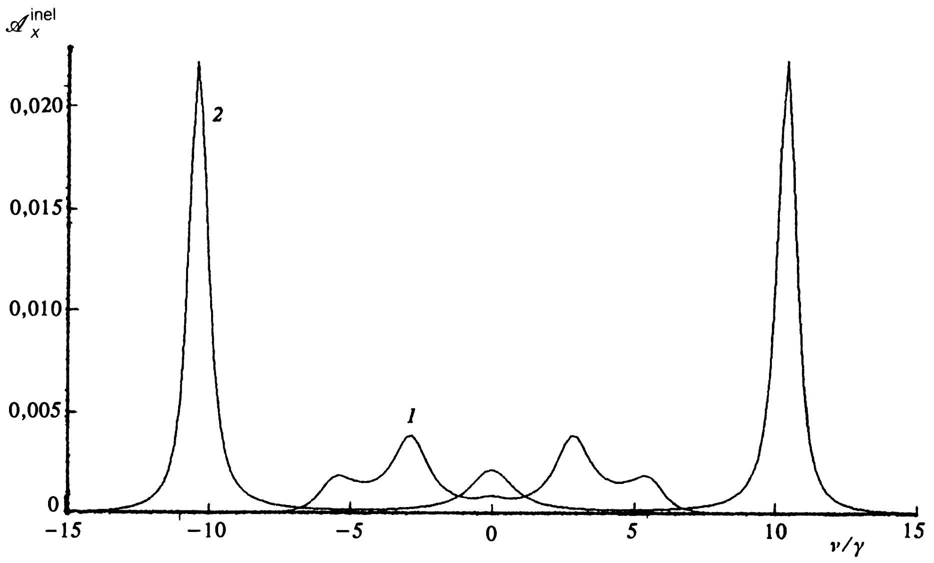

FIG. 4. Spectral dependence of the inelastic component of the Hanle signal for $\Delta_L=0$, $V/\gamma=2$, and $\Omega/\gamma=0.2$ (*1*), 10 (*2*).

pressions for the poles of the retarded Green's functions.[9,14] It is found by solving the cubic equation[9]

$$\varepsilon(\varepsilon-\Delta_+)(\varepsilon-\Delta_-)-|V_+|^2(\varepsilon-\Delta_-)$$
$$-|V_-|^2(\varepsilon-\Delta_+)=0. \quad (25)$$

For V_+, V_-, $\Omega \gg \Delta$ we find an approximate solution of this equation:

$$\varepsilon_0=\Delta\left(1+\frac{\Omega^2}{3W^2}\right),\quad \varepsilon_\pm=\pm W+\frac{\Delta}{2}\left(1-\frac{\Omega^2}{3W^2}\right), \quad (26)$$

where $W=(|V_+|^2+|V_-|^2+\Omega^2)^{1/2}$, $\Delta=\Delta_L-i\gamma/2$.

A state diagram of the dressed atom of the system under consideration is shown in Fig. 6. From the diagram it is clear that seven lines should be visible in the spectrum of the scattered radiation since three of the ten transitions are spectrally degenerate. Note in this case that the spectrum of transitions is determined by the real part of the eigenvalues of the matrix **M**. It follows from expressions (26) that when the laser wave is in exact resonance with the transition frequency ($\Delta_L=0$) the quasi-energy spectrum becomes equidistant and it should be possible to observe five lines in the emission spectrum. This is in agreement with the fluorescence spectra in Figs. 3–5.

Despite the cumbersome form of the spectrum of the Hanle signal, which is given by Eqs. (16), (10), and (12), in some limiting cases relatively simple expressions can be obtained.

For the case of very strong magnetic fields $\Omega \gg V, \gamma, \Delta_L$ the spectrum of the inelastic component consists mainly of two peaks and can be approximated in those regions where it is significant by a superposition of Lorentzians:

$$\mathscr{A}_x^{\text{inel}} \approx Ng^2\frac{4V^4}{\Omega^4}\left[\frac{\gamma}{(\nu-\Omega)^2+\gamma^2/4}+\frac{\gamma}{(\nu+\Omega)^2+\gamma^2/4}\right]. \quad (27)$$

Note that, as in the Mollow spectrum, the Hanle signal in

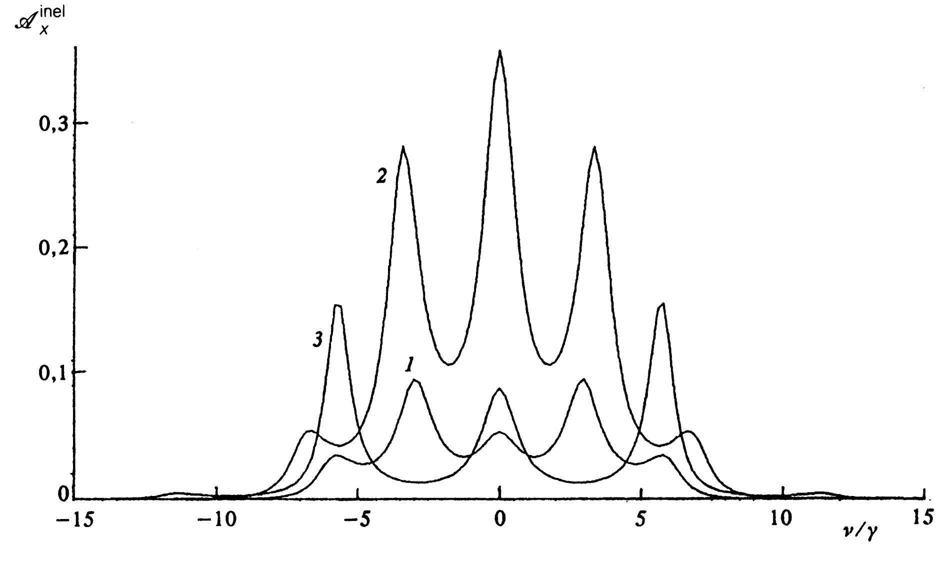

FIG. 5. Spectral dependence of the inelastic component of the Hanle signal for $\Delta_L=0$, $V/\gamma=2$, and $\Omega/\gamma=1$ (*1*), 2 (*2*), 5 (*3*).

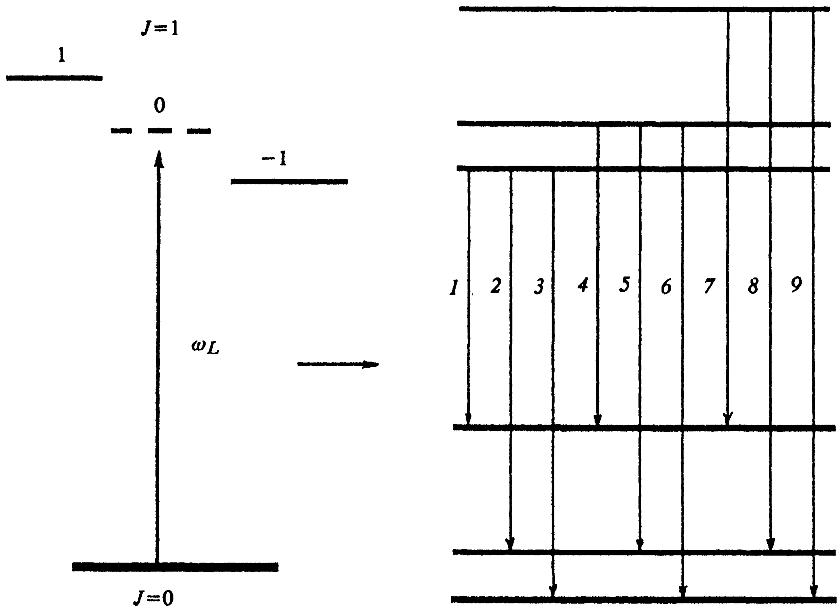

FIG. 6. Energy level diagram of the $\mathbf{J}=0\rightarrow\mathbf{J}=1$ atomic system in a constant magnetic field under the action of a strong laser wave, showing shift and splitting. The lines corresponding to transitions *3*, *5*, and *7* are spectrally degenerate with the frequency of the laser line.

weak fields is proportional to V^4 (the elastic component in this case is proportional to V^2) and expression (27) is also valid for $V \gg \gamma$, but only when $V \ll \Omega$. Comparing these results with the fluorescence spectra of the two-level atom,[8] we can easily see that the Larmor frequency in this case is analogous to the large detuning of the radiation from resonance for a two-level atom.

Let us consider the limit in which the intensity of the incident wave is very high, when V is much greater than Ω and γ for the case of exact resonance $\Delta_L=0$. In this case the spectrum of the Hanle signal consists mainly of four side components and can be approximated in the following way:

$$\mathscr{A}_x^{\text{inel}} \approx N g^2 \frac{\Omega^2}{2V^2}\left[\frac{3\gamma/8}{(\nu-2W)^2+9\gamma^2/16} + \frac{3\gamma/4}{(\nu-W)^2+9\gamma^2/16} + \frac{3\gamma/4}{(\nu+W)^2+9\gamma^2/16} + \frac{3\gamma/8}{(\nu+2W)^2+9\gamma^2/16}\right]. \tag{28}$$

It can be seen from this expression that the height of the inner peaks is twice that of the outer ones. As in the Mollow spectrum for this case, the widths of the side components are one and a half times larger than the width of the central component, which in this case is small since its contribution is inversely proportional to V^4.

5. CONCLUSION

Our study of the spectral features of the Hanle effect for the monochromatically excited $\mathbf{J}=0\rightarrow\mathbf{J}=1$ atomic system has shown that, as in the fluorescence spectrum of a two-level atom, the spectrum of the Hanle signal contains an elastic and an inelastic component. There can be as many as seven peaks in the spectrum of the shifted component. In intense electromagnetic fields the shifted component predominates, but this requires that the magnetic fields not be too strong: the Larmor frequency must be smaller than the Rabi frequency. For a weak incident wave or in very strong magnetic fields the scattering becomes Rayleigh.

It should be noted that our analysis is correct only for optically thin media, i.e., it does not take absorption of the scattered radiation into account. In optically thick media it is necessary to consider the entire system of equations (8), (9), not just their free terms.

The resonance fluorescence spectra for a strong monochromatic wave have been well investigated experimentally.[15–17] Recently, Zhu *et al.*[18] have observed resonance fluorescence with a multipeak structure by irradiating atoms with intense biharmonic radiation. An experimental study of the spectral features of the Hanle effect for a strong monochromatic wave is a technically similar problem, and therefore should be feasible.

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