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The Hanle effect in a strong electromagnetic field

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A theory of the Hanle effect in a strong electromagnetic field, when perturbation theory is inapplicable, is developed. A two-level system with terms of which one has zero and the other unity angular momentum is considered. The probability for transition to a third level with zero angular momentum under conditions when the first two terms are at resonance with the strong field is computed. It is shown that the dependence of the probability changes from quadratic to linear as the field intensity is increased. The limits of very weak and very strong fields and the case of a very strong constant magnetic field that splits the term with unity angular momentum are analytically investigated. The intermediate cases are investigated with the aid of a computer calculation. The self-similar character of the problem is pointed out. It is concluded that the resonant character of the probability as a function of the magnetic field vanishes as the electric-field intensity increases.

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§1. FORMULATION OF THE PROBLEM

The optical phenomena connected with interference, due to the presence of adjacent levels, in the radiation of atoms are currently being intensively investigated.[1] One of these effects is the Hanle effect, which consists in the fact that in a magnetic field the intensity of spontaneous radiation with a given polarization depends on the distance, determined by the magnetic-field strength, between the adjacent Zeeman sublevels. The Hanle effect is explained by the fact that the probability of emission of radiation with some definite polarization for an atomic state that is a superposition of energetically close states is determined by the square of the modulus of the sum of the occupation amplitudes of these states. The dependence of the probability on the level spacing is due to the presence of an interference term in the square of the modulus of the sum.

The Hanle effect is normally observed in resonance excitations by radiation with a broad spectral line. Furthermore, it occurs in resonance excitations by monochromatic light.[2] In this case the Hanle signal depends not only on the distance between the adjacent levels, but also on the detunings of the resonances with the field frequency. But, as a rule, an ensemble of absorbing atoms has, as a result of the Doppler effect, a fairly broad resonance-detuning distribution function. For this reason, the averaged shape of the Hanle signal does not depend on the resonance detunings, and is equivalent to the shape that is attained in excitations by radiation with a broad spectral line. This assertion is valid only for low intensities of the monochromatic light, when perturbation theory is applicable.[2]

In the present paper we consider the Hanle effect in a strong electromagnetic field. The process proceeds as follows (see Fig. 1). The initial atomic state a (having a radiative width γ_a) is populated with the aid of some external field, V_0, of a high-power pulsed laser. The time during which the field V_0 acts is assumed to be short compared to all the characteristic times of the problem. The population of the adjacent levels b and b' from the state a is attained as a result of the action of a strong resonance monochromatic external field, $2v\cos\omega t$, of a cw laser. The states b and b' possess radiative widths of γ_b and $\gamma_{b'}$, which, following Chaika[1] and Series,[2] we shall assume below to be equal: $\gamma_b = \gamma_{b'} \equiv \gamma$. The main difference between the present paper and Series's paper[2] is that we do not use perturbation theory in terms of v.

The level diagram shown in Fig. 1 actually implies that we are considering the case in which the state a has zero and the state b unity angular momentum. In a magnetic field, the state b splits into three states, b, b', and b'', with magnetic quantum numbers m, i.e., with angular-momentum components along the direction of the magnetic field equal respectively to $+1$, -1, and 0. If the incident radiation is linearly polarized along a direction perpendicular to the direction of the magnetic field (say along the x axis if the magnetic field is directed along the z axis), then the matrix elements for the transitions $0\to+1$ and $0\to-1$ are nonzero, i.e., the state b'' does not participate in the process under consideration. For other angular-momenta of the levels a large number of states is drawn into the problem and complicates it quantitatively. It is easy to see that, for incident radiation polarized along the x axis, the matrix elements of the $a\to b$ and $a\to b'$ transitions will be equal, i.e., $v_{ab}=v_{ab'}\equiv V$ while in the case of polarization along the y axis $v_{ab}=-v_{ab'}=iV$.

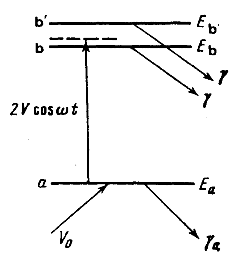

FIG. 1. Level diagram for the investigation of the Hanle effect. The levels b and b' are close to each other. The level a is populated upon excitation by a pulsed field V_0. The level a and the levels b and b' are mixed by an external resonant field of frequency ω.

We compute the probability for the population of some zero-angular-momentum level, c, as a result of spontaneous transitions to it from the adjacent terms b and b', which are mixed by the field V. This is accompanied by emission of a photon with some frequency ν. The distribution over the frequencies ν is normally of no interest in measurements, and therefore the computed probability should be integrated over ν. The result depends on the distance between the adjacent levels b and b' and on the resonance detunings $\omega_{ba}-\omega$ and $\omega_{b'a}-\omega$. But, as in Ref. 2, we assume that an ensemble of absorbing atoms has, on account of the Doppler effect, a sufficiently broad detuning distribution. The width of this distribution is assumed to be large compared to both the distance between the adjacent levels and the spontaneous widths γ and γ_a. Since the motions of the individual atoms in the gas are not correlated, the probability for a spontaneous transition to the level c should be further averaged over the atomic velocities.

For a strong $2v\cos\omega t$ field, the method that allows us to go outside the framework of perturbation theory is the resonance-approximation method. Thus, we assume that the field frequency ω is close to ω_{ba} and $\omega_{b'a}$. In this case it is possible for V to be greater or smaller than $\omega_{b'b}$, but we should, of course, have $V \ll \omega$.

The case of the Hanle effect in a strong electromagnetic field, $2v\cos\omega t$, has been considered also by Ducloy.[3,4] In contrast to our present formulation of the problem, Ducloy[3,4] assumed that the action time of the field V_0 that populates the initial state a is long compared to all the characteristic times of the problem. This allowed him to limit himself to the investigation of the ordinary steady state in the equations describing the Hanle effect. But in this regime the population of the level a is small, on account of the spontaneous decay of the level, and this sharply decreases the Hanle signal. Unlike Ducloy,[3,4] we consider the case in which the level a is pumped by ultrashort high-power laser pulses.

Section 2 contains the equations of the theory and the method of solution. In §3 we present the results of a numerical computer solution of these equations, as well as various analytical limiting cases.

§2. THE EQUATIONS AND THEIR GENERAL SOLUTION

Let us write down, in accordance with the notation introduced in Fig. 1, the system of equations describing the transitions between the states a, b, and b' in the resonance approximation. We shall, on the basis of the Breit–Wigner procedure, take the level widths into account by adding imaginary corrections to the corresponding energies.

Let us denote by $a(t)$, $b(t)$, and $b'(t)$ the amplitudes for the population of the respective states. The levels b and b' are close to each other. They are, from the experimental point of view, states with magnetic quantum numbers $+1$ and -1 respectively, and upon the application of a constant magnetic field the distance between them changes as a result of the Zeeman effect.

At the initial moment of time $t=0$, an electron occupies the state a as a result of the action of a high-power pulsed laser field V_0. A continuously acting resonance laser field, $2v\cos\omega t$, is applied to the system.

The equations for the amplitudes $a(t)$, $b(t)$, and $b'(t)$ have the form

$$\begin{aligned} i\dot{a}&=v_{ab}\exp(-i\delta_{ba}t)\,b+v_{ab'}\exp(-i\delta_{b'a}t)\,b',\\ i\dot{b}&=v_{ba}\exp(i\delta_{ba}t)\,a,\\ i\dot{b}'&=v_{b'a}\exp(i\delta_{b'a}t)\,a. \end{aligned} \tag{1}$$

Here the quantities

$$\delta_{ba}=\omega_{ba}-\omega-{}^1/_2i(\gamma-\gamma_a),$$
$$\delta_{b'a}=\omega_{b'a}-\omega-{}^1/_2i(\gamma-\gamma_a)$$

are the detunings of the respective resonances with allowance for the spontaneous level widths.

We seek the basis solutions to the system of equations (1) in the standard form

$$\begin{aligned} a&=A\exp(i\Omega t),\\ b&=B\exp[i(\Omega+\delta_{ba})t],\\ b'&=B'\exp[i(\Omega+\delta_{b'a})t]. \end{aligned} \tag{2}$$

The quantity Ω can be called the Rabi frequency for the problem under consideration,[5] or a quasienergy. For Ω we obtain from (1) and (2) the cubic equation

$$\Omega(\Omega+\delta_{ba})(\Omega+\delta_{b'a})-V^2(2\Omega+\delta_{ba}+\delta_{b'a})=0. \tag{3}$$

Equation (3) has three roots, Ω_1, Ω_2, and Ω_3, and determines three corresponding sets of coefficients, A_i, B_i, and B_i' $(i=1, 2, 3)$. Thus, we find that the general solution to the problem is a superposition of the three indicated basis solutions. The three constants in this superposition are found from the initial conditions $a(0)=1$ and $b(0)=b'(0)=0$. By the same token the problem of finding the wave function of the system is, in principle, solved completely. It is easy to verify from the system (1) that the law of particle-number conservation,

$$\gamma_a\int_0^\infty|a(t)|^2\exp(-\gamma_a t)\,dt+\gamma\int_0^\infty[|b(t)|^2+|b'(t)|^2]\exp(-\gamma t)\,dt=1,$$

which manifests itself in the fact that the particle is totally absent from the system in question during the entire action time of the perturbation, is satisfied. Here it is assumed that the spontaneous-decay channels for the a, b, and b' states are different and independent of each other; only then will the Breit–Wigner procedure be valid.[6]

The probability for radiative transition from the states b and b', which are mixed by the field V, to a final state c with a definite polarization takes, after being integrated over the frequencies ν of the emitted photons, the following form:

$$w(\omega)=\gamma_{bc}\int_0^\infty|b(t)\exp(-iE_bt)\pm b'(t)\exp(-iE_{b'}t)|^2\exp(-\gamma t)\,dt. \tag{4}$$

In Eq. (4) the plus sign corresponds to a photon emitted with polarization along the x axis; the minus sign, to a photon emitted with polarization along the y axis. Further, γ_{bc} is the probability per unit time for spontaneous transition from the state b to the state c with polarization along the x or y axis (and is equal to the quantity $\gamma_{b'c}$).

Equation (4) is applicable when the condition $c\neq a$ is fulfilled. In the opposite case the Breit–Wigner procedure used above to write down the system of equations (1) is incorrect, since the channel c exerts considerable reaction on the population of the levels b and b', as a result of which the problem becomes quite complicated.

Notice that in the case of a weak field the Hanle effect is due to the interference term of the probability $w(\omega)$ given by Eq. (4).

Because of the Doppler effect, the quantity $w(\omega)$ depends on the atomic velocity $\mathbf{v}$ through the combination $\omega\to\omega+\mathbf{k}\cdot\mathbf{v}$, where $\mathbf{k}$ is the wave vector. The averaging of the probability w over the velocity $\mathbf{v}$ amounts to integration over the values of ω. Thus, we have

$$\bar{w}=\int_{-\infty}^{\infty}w(\omega)\,d\omega. \tag{5}$$

The integration limits can be extended here to infinity because of the large Doppler width, since the integrand is concentrated in the narrow region $\omega\approx\omega_{ba}$, $\omega_{b'a}$, and a narrow strip is actually cut out from the Doppler contour.

§3. LIMITING CASES AND NUMERICAL RESULTS

The quantity $\bar{w}$ has an analytical form only in the various limiting cases, since in the general case the solutions to Eq. (3) are given by unwieldy Cardano formulas, as a result of which the integration in the formula (5) can be performed only by numerical methods.

Let us first consider the well-known case of perturbation theory. It is realized in weak fields, namely, when the conditions

$$V\ll\gamma,\ \gamma_a$$

are fulfilled. Then we obtain for the probability $\bar{w}$ from (4) and (5) the following expression[1,2]

$$\bar{w}=\mathrm{const}\,V^2\left[1\pm\frac{\gamma^2}{\omega_{b'b}^2+\gamma^2}\right]. \tag{6}$$

The plus sign in this formula corresponds to the case in which the polarizations of the incident and scattered radiations are the same (and perpendicular to the polarization of the magnetic field), while the minus sign corresponds to the case in which the polarization of the emitted photons is perpendicular to that of the incident radiation (and both of them are also perpendicular to the polarization of the magnetic field).

The first term in Eq. (6) stems from the squares of the moduli of the amplitudes $b(t)$ and $b'(t)$ in (4), while the second term is connected with the interference term in (4). It has a resonance structure, and describes the Hanle effect proper. As we can see, the resonance width is determined by the b- and b'-level widths. The nonresonant terms do not depend on this width, which is natural, since b and b' are intermediate levels.

Let us now consider another limiting case, which corresponds to the case in which the levels b and b' are very close to each other. In this case the expression (5) can be analytically integrated only when the levels b, b', and a have the same width, i.e., when $\gamma_a = \gamma$. Assuming that this condition is fulfilled, we find from (4) and (5) that when the incident and observed radiations have the same polarization

$$\bar{w} = \text{const}\, V^2/(\gamma^2+8V^2)^{1/2}, \qquad (7)$$

while in the case in which the polarizations are perpendicular to each other the probability is equal to zero. The expression (7) is applicable if

$$\gamma^2+8V^2 \gg \omega_{b'b}^2.$$

It is easy to see that in the parameter region where $V \ll \gamma$ the expression (7) has the same form as the expression (6) in the parameter region where $\omega_{b'b} \ll \gamma$, as expected on the basis of the simulataneous applicability of the solutions in this region.

Finally, the following analytical solution is obtained upon the fulfillment of the conditions

$$V, \gamma \ll \omega_{b'b}.$$

These conditions correspond to the situation in which there is no interference between the levels because of the great distance between them. In this case the two-step transitions from a to c via the states b and b' occur independently of each other. Then from Eqs. (4) and (5) we find

$$\bar{w} = \text{const}\, V^2/(\gamma^2+4V^2)^{1/2}. \qquad (8)$$

Notice that the expressions (6) and (8) coincide when the conditions

$$V \ll \gamma \ll \omega_{b'b}$$

are fulfilled, as expected from the conditions for their simulataneous applicability.

In Fig. 2 we show the regions defined by the variables V/γ and $\omega_{b'b}/\gamma$ where the solution is given by one of the analytical formulas (6), (7), and (8). We see that these regions overlap partially.

Numerical computations are necessary for the intermediate parameter values. Figure 3 shows the results of such computations for the case in which the incident and observed radiations are identically polarized in a direction perpendicular to the polarization of the magnetic field and $\gamma_a = \gamma$. The ordinate in this figure is the probability $\bar{w}$ in units of $w_0 = 2\pi V^2 \gamma_{bc}/\gamma$.

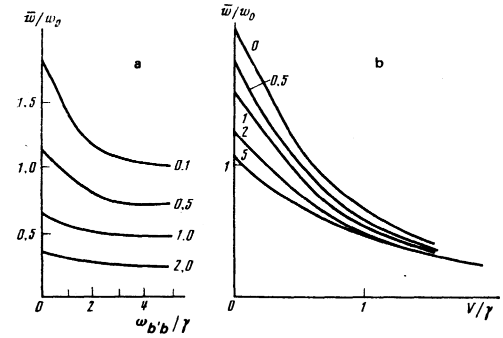

FIG. 3. a) Dependence of the radiative-transition probability on the distance $\omega_{b'b}$ between the adjacent terms, in units of γ, for a fixed value of the perturbation amplitude V expressed in units of γ (the value is indicated near each curve). b) Dependence of the radiative-transition probability on the perturbation amplitude V for a fixed distance $\omega_{b'b}$ between the levels (this distance is indicated near each curve); all the quantities are expressed in units of γ.

It can be seen from Fig. 3a that the resonant structure of the probability gets gradually smoothed out as the field strength V is increased. The locations of the peaks in this figure are given by the formula (7); as the quantity $\omega_{b'b}/\gamma$ increases, the curves approach asymptotic values that are independent of $\omega_{b'b}$ and are given by Eq. (8). This smoothing can be attributed to the mutual repulsion of the levels b and b' in a strong field. As a result of this repulsion, the overlap of these levels decreases, on account of their broadening of γ, which implies the weakening of the interference, i.e., of the Hanle effect.

Let us note that for strong fields we should expect a linear dependence of the probability on the incident-radiation intensity, while in weak fields this dependence is, according to (6), quadratic. This result can be seen from the formulas (7) and (8) if we set $V \gg \gamma$.

In the present paper we have considered the case of two adjacent levels. We have found that a strong field alters significantly the shape of the Hanle signal. Similar effects, which can be analyzed by a similar method, are to be expected when the number of adjacent levels is increased, i.e., when we go over to states with other angular momenta.

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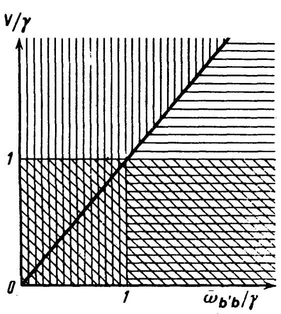

FIG. 2. The plane of the variables V/γ and $\omega_{b'b}/\gamma$, in which analytical solutions to the Hanle problem exist. The obliquely hatched area corresponds to the region in which perturbation theory is applicable [Eq. (6)]; the vertically hatched area corresponds to the region of very close terms [Eq. (7)]; the horizontally hatched area corresponds to the region in which the terms are far apart [Eq. (9)].

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Two-photon excitation of a quantum system

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To analyze the crossing of the quasienergy levels of a system interacting with an intense alternating field under conditions of two-photon resonance, we propose an exactly solvable model of a field whose envelope is of the characteristic interaction switching-on type. The kinetics of the system in the field is analyzed. From the obtained general relation for the probability of two-photon excitation there follow as limits instantaneous switching-on of the field and the adiabatic limit (the Landau-Zener formulas). The dependence of the excitation probability on the field intensity and the detuning of the two-photon resonance is analyzed for different interaction switching-on regimes and with allowance for possible ionization of the system from the upper state.

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1. INTRODUCTION. FORMULATION OF THE PROBLEM

Two-photon excitation is one of the first experimentally observed effects of nonlinear optics.[1] A large number of studies have now been made in which two-photon excitation has been observed in both condensed media and in gases. The theoretical description of the probability of two-photon excitation of state 2 from state 1 is usually based on the Weisskopf–Wigner formula, which also describes single-photon excitation[2]:

$$W=|V_{12}|^2\frac{\Gamma}{(E_1-E_2+2\omega)^2+\Gamma^2/4}. \qquad (1)$$

Here, V_{12} is the matrix element of the two-photon transition, E_1 and E_2 are the energies of the levels, Γ is the homogeneous line width, $\hbar=1$, and absence of saturation is also assumed: $|V_{12}|\ll|E_2-E_1-2\omega+i\Gamma/2|$. In the presence of inhomogeneous broadening, expression (1) must be appropriately averaged.

In the absence of intermediate single-photon resonance, which will be assumed in what follows, the matrix element V_{12} depends linearly on the radiation intensity I. The energy levels $E_{1,2}$ also depend linearly on the intensity because of the quadratic dynamical Stark effect.[1)] This fact can be taken into account by setting in formula (1)

$$E_{1,2}=E_{1,2}^{(0)}-\alpha_{1,2}I/4, \qquad (2)$$

where $E_{1,2}^{(0)}$ are the energy levels in the absence of radiation, and $\alpha_{1,2}$ are the polarizabilities of the levels at the field frequency ω.

Equations (1) and (2) can be proved by means of Low's equations, which describe the natural width of atomic levels if one takes into account the contribution to the mass operator of not only the photon vacuum but also the field of the laser radiation.[4] For this, however, it is necessary to assume that the electromagnetic field is stationary, for otherwise the mass operator, which is a function of two four-points, becomes dependent on t and t' separately and not merely on the difference $t-t'$, and as a result Eq. (1) cannot be proved.

Since there cannot be strictly stationary laser fields (if only because of the existence of the switching-on period), formula (1) is by no means always valid. Indeed, in recent studies[5-8] it was shown that in a number of cases two-photon excitation bears a greater similarity to the transitions between molecular terms in slow collisions of atoms or adiabatic spin inversion in magnetic resonance than to the resonant absorption of a single photon.

Figure 1 explains the physical situation. Suppose, for simplicity, that the time dependence of the radiation intensity is due solely to the switching-on of the field. Then the energy levels $E_{1,2}(t)$ vary from $E_{1,2}^0$ to certain stationary values determined by the steady-state field intensity, as shown in Fig. 1. At definite values of the detuning from resonance in the absence of radiation, of the difference between the level polarizabilities, and of the intensity in the steady state, it is possible for the levels $E_1+2\omega$ and E_2 to cross at a certain time t_0. As is well known,[9] the presence of even weak interaction between states 1 and 2 leads to quasi-

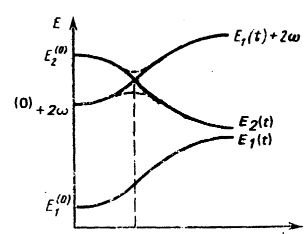

FIG. 1.

0038-5646/81/020241-07$02.40