ON EVALUATING THE POLARIZATION OF AN ATOM IN A STRONG ELECTROMAGNETIC FIELD

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Submitted to JETP editor June 12, 1965

J. Exptl. Theoret. Phys. (U.S.S.R.) 49, 1605-1610 (November, 1965)

The interaction of a two-level quantum system with the sum of a weak and strong monochromatic radiation field is calculated. The conditions are obtained for which the polarization components at the combination frequencies are of the same order of magnitude as the components at the weak field frequencies.

THERE is much current interest in the problem of the interaction of a two-level quantum system with a strong radiation field in relation to a number of questions arising in the theory of quantum generators. This problem has been solved for a monochromatic field by Karplus and Schwinger [1]. Lamb [2] discussed the influence of fields of several frequencies upon the system, but the fields were assumed to be weak. Sobel'man and one of the authors [3] first considered the case of two monochromatic fields—weak and strong—and calculated the amplification factor for the weak field. However, polarization components at the combination frequencies also occur as a result of the influence of strong and weak monochromatic fields upon the atom. Therefore it is not possible to introduce the concept of amplification factor in the case of a nonmonochromatic weak field. In the present work we calculate the polarization of an atom due to a strong monochromatic field and an arbitrary weak field. Furthermore, we examine a more general two-level system (by comparison with [3]), for which the width of the fluorescence line differs from the sum of the population relaxation constants.

Let us write the Hamiltonian of the system in the form

$$H = H_0 - \hat{p}E,$$

where H_0 is the Hamiltonian of the system without the perturbing field, $\hat{p}$ is the operator of the dipole moment, and E is the electric field strength. If the field effectively interacts only with two levels of the operator H_0, it is possible to calculate the polarization by introducing the second order density matrix. Subsequently, following Lamb[2], we shall always consider the density matrix averaged over the excitation moments. The elements of the averaged density matrix ρ are described by the system of equations:

$$\frac{d}{dt}\rho_{11} + 2\gamma_1\rho_{11} = -i\frac{p}{\hbar}E(\rho_{12} - \rho_{12}^*) + \Lambda_1,$$
$$\frac{d}{dt}\rho_{22} + 2\gamma_2\rho_{22} = i\frac{p}{\hbar}E(\rho_{12} - \rho_{12}^*) + \Lambda_2,$$
$$\frac{d}{dt}\rho_{12} + (\tilde{\Gamma} - i\omega_0)\rho_{12} = i\frac{p}{\hbar}E(\rho_{22} - \rho_{11}). \quad (1)$$

Here the indices 2 and 1 refer to the upper and lower levels, respectively; Λ_2 and Λ_1 are the excitation rates of the levels being studied, and ω_0 is the characteristic frequency of the transition between them; the matrix elements of the dipole moment are $p_{11} = p_{22} = 0$, $p_{12} = p$, and it can be assumed that $p = p^*$ and $p > 0$. The system is characterized by the three relaxation constants γ_1, γ_2, and Γ.

We note that in the literature on quantum generators equations for a two-level system are often seen which, unlike Eq. (1), satisfy the condition that the sum of populations in levels 1 and 2 is constant. For example, equations such as that below are frequently encountered:

$$\frac{d}{dt}\rho_{11} + \Gamma_1\rho_{11} - \Gamma_2\rho_{22} = -i\frac{p}{\hbar}E(\rho_{12} - \rho_{12}^*),$$
$$\frac{d}{dt}\rho_{22} + \Gamma_2\rho_{22} - \Gamma_1\rho_{11} = i\frac{p}{\hbar}E(\rho_{12} - \rho_{12}^*),$$
$$\frac{d}{dt}\rho_{12} + (\tilde{\Gamma} - i\omega_0)\rho_{12} = i\frac{p}{\hbar}E(\rho_{22} - \rho_{11}). \quad (2)$$

Our calculation will be applicable to such systems as well. The population difference and polarization for Eqs. (2) can be obtained by setting $\Lambda = \Gamma_1 - \Gamma_2$, $\Gamma = \Gamma_1 + \Gamma_2$, and $\gamma = 0$ in the final formulae (9) and (11). Henceforth, we shall discuss only the solution of system (1).

Let us first consider the case where there are both a strong and a weak monochromatic field:

$$E = \frac{\hbar}{p} F \cos \omega t + \frac{\hbar}{p} f \cos(\omega t + \Omega t + \varphi). \quad (3)$$

The first term of (3) is assumed to be the strong field, and the second the weak field. It is assumed that the frequencies ω and $\omega + \Omega$ are close to the transition frequency ω_0.

Let us transform to new unknown real variables y_n:

$$\rho_{11} + \rho_{22} = y_1; \quad \rho_{22} - \rho_{11} = y_2; \quad \rho_{12} = e^{i\omega t}(y_3 + iy_4).$$

In addition, we introduce the notation

$$\Gamma = \gamma_1 + \gamma_2, \quad \gamma = \gamma_1 - \gamma_2,$$

$$\Lambda = \Lambda_2 - \Lambda_1, \quad \lambda = \Lambda_1 + \Lambda_2. \quad (4)$$

Using (3) and (4), we obtain from (1) a system of equations for y_n:

$$\frac{d}{dt} y_1 + \Gamma y_1 = \gamma y_2 + \lambda, \quad \frac{d}{dt} y_2 + \Gamma y_2 = \gamma y_1 + \Lambda$$

$$+ 2i [F \cos \omega t + f \cos(\omega t + \Omega t + \varphi)]$$

$$\times [(y_3 + iy_4) e^{i\omega t} - (y_3 - iy_4) e^{-i\omega t}],$$

$$\frac{d}{dt}(y_3 + iy_4) + (\tilde{\Gamma} + i\omega - i\omega_0)(y_3 + iy_4)$$

$$= ie^{-i\omega t} [F \cos \omega t + f \cos(\omega t + \Omega t + \varphi)] y_2. \quad (5)$$

Further (following the authors of [1-3]), we discard from the right sides of (5) the terms containing the frequency 2ω:

$$\frac{d}{dt} y_1 + \Gamma y_1 = \gamma y_2 + \lambda, \quad \frac{d}{dt} y_2 + \Gamma y_2 = \gamma y_1 + \Lambda - 2F y_4$$

$$+ 2f y_3 \sin(\Omega t + \varphi) - 2f y_4 \cos(\Omega t + \varphi), \quad (6)$$

$$\frac{d}{dt}(y_3 + iy_4) + (\tilde{\Gamma} + i\omega - i\omega_0)(y_3 + iy_4)$$

$$= \frac{i}{2}(F + f e^{i\Omega t + i\varphi}) y_2.$$

We seek the solution of the system (6) in the form of an exact solution of this system for $f = 0$ and corrections that depend upon the weak field

$$y_n = X_n + x_n.$$

The zeroth-approximation equations are of the form

$$\frac{d}{dt} X_1 + \Gamma X_1 - \gamma X_2 = \lambda,$$

$$\frac{d}{dt} X_2 + \Gamma X_2 - \gamma X_1 + 2F X_4 = \Lambda,$$

$$\frac{d}{dt}(X_3 + iX_4) + (\tilde{\Gamma} + i\omega - i\omega_0)(X_3 + iX_4) - \frac{i}{2} F X_2 = 0 \quad (7)$$

and are well known in the literature; for the corrections x_n we obtain, discarding quantities of the type fx_n, the following system of equations:

$$\frac{d}{dt} x_1 + \Gamma x_1 - \gamma x_2 = 0, \quad \frac{d}{dt} x_2 + \Gamma x_2 - \gamma x_1 + 2F x_4$$

$$= 2f X_3 \sin(\Omega t + \varphi) - 2f X_4 \cos(\Omega t + \varphi),$$

$$\frac{d}{dt}(x_3 + ix_4) + (\tilde{\Gamma} + i\omega - i\omega_0)(x_3 + ix_4) - \frac{i}{2} F x_2$$

$$= \frac{i}{2} f X_2 e^{i\Omega t + i\varphi}. \quad (8)$$

It is not difficult to verify that the homogeneous system of equations corresponding to (7) and (8) has a solution only for complex frequencies, i.e., unbounded at $t = \pm\infty$. Therefore the solutions of the inhomogeneous systems (7) and (8) satisfying the boundedness requirement within $-\infty < t < +\infty$, are determined uniquely.

The solution of (7) is of the form

$$X_2 = \left(\frac{\Lambda}{\Gamma} + \frac{\lambda\gamma}{\Gamma^2}\right) \Big/ \left\{1 - \frac{\gamma^2}{\Gamma^2} + \frac{F^2}{\Gamma\tilde{\Gamma}}\left[1 + \left(\frac{\omega - \omega_0}{\tilde{\Gamma}}\right)^2\right]^{-1}\right\},$$

$$X_3 + iX_4 = \frac{i}{2} \frac{F}{\tilde{\Gamma} + i(\omega - \omega_0)} X_2. \quad (9)$$

The system (8) is best solved using the substitution

$$x_n = \mathrm{Re}(z_n e^{i\Omega t + i\varphi}).$$

We obtain from (8) a system of linear algebraic equations in z_n:

$$(i\Omega + \Gamma) z_1 - \gamma z_2 = 0, \quad (i\Omega + \Gamma) z_2 - \gamma z_1 + 2F z_4$$

$$= -2f(X_3 + iX_4), \quad (i\Omega + \tilde{\Gamma}) z_3 - (\omega - \omega_0) z_4 = \frac{i}{2} f X_2,$$

$$(i\Omega + \tilde{\Gamma}) z_4 + (\omega - \omega_0) z_3 - \frac{1}{2} F z_2 = \frac{1}{2} f X_2. \quad (10)$$

Thus, solving (10), we find

$$z_2 = -fFX_2 \frac{2\tilde{\Gamma} + i\Omega}{\tilde{\Gamma} + i\Omega} \frac{\tilde{\Gamma} - i(\omega - \omega_0 - \Omega)}{\tilde{\Gamma} - i(\omega - \omega_0)} \left\{\left[i\Omega + \tilde{\Gamma} + \frac{(\omega - \omega_0)^2}{i\Omega + \tilde{\Gamma}}\right]\left[i\Omega + \Gamma - \frac{\gamma^2}{i\Omega + \Gamma}\right] + F^2\right\}^{-1}, \quad (11)$$

$$z_3 + iz_4 = \frac{ifX_2}{\tilde{\Gamma} + i(\omega - \omega_0 + \Omega)} + \frac{iFz_2/2}{\tilde{\Gamma} + i(\omega - \omega_0 + \Omega)}, \quad (12)$$

$$z_3^* + iz_4^* = \frac{iFz_2^*/2}{\tilde{\Gamma} + i(\omega - \omega_0 - \Omega)}. \quad (13)$$

The average value of the dipole moment (or the polarization per particle) is expressed by means of the quantities which we have calculated in the following form:

$$p_{\text{av}} = p\,\mathrm{Re}\,[2(X_3 + iX_4)e^{i\omega t} + (z_3 + iz_4)e^{i(\omega t + \Omega t + \varphi)} + (z_3^* + iz_4^*)e^{i(\omega t - \Omega t - \varphi)}]. \quad (14)$$

Thus, in the approximation considered, in addition to components at the frequencies ω and $\omega + \Omega$ present in the original electromagnetic field, there also appears a component at the combination frequency $\omega - \Omega$.

Let us examine Eq. (14) in greater detail. The quantity $X_3 + iX_4$ determines the polarization component at the strong-field frequency, and is given [see (9)] by the product of the strong-field amplitude F, the population difference X_2, and the resonance factor $[\tilde{\Gamma} + i(\omega - \omega_0)]^{-1}$. The weak-field-frequency polarization component is determined by the quantity $z_3 + iz_4$. It can be seen from (12) that $z_3 + iz_4$ consists of two terms. The first is quite simple in form and is identical to the quantity which determines the strong field polarization, except that F is replaced by f and the resonance factor $[\tilde{\Gamma} + i(\omega - \omega_0)]^{-1}$ by $[\tilde{\Gamma} + i(\omega + \Omega - \omega_0)]^{-1}$. If F approaches zero, the first term in $z_3 + iz_4$ remains finite.

The second term in $z_3 + iz_4$ is more complicated. The F-dependence of the second term does not reduce simply to the effect of the strong field F upon the population difference. This is obvious from Eq. (11) for z_2 wherein, in addition to the population difference X_2, we have an additional F-dependent factor. As $F \to 0$, z_2 and hence the entire term vanishes. The frequency dependence of this term is contained both in the factor $[\tilde{\Gamma} + i(\omega + \Omega - \omega_0)]^{-1}$ and in z_2.

The quantity $z_3^* + iz_4^*$ in (14) describes the polarization component at the frequency $\omega - \Omega$. This frequency was not contained in the original field, and its appearance is evidence of the nonlinear properties of the system in a strong radiation field. It can be seen from (12) and (13) that the modulus of $z_3^* + iz_4^*$ and the modulus of the second term in the expression for $z_3 + iz_4$ are of the same order of magnitude. This means that the appearance of the combination frequency in the polarization will occur in all cases when the deviation of the frequency dependence of the amplification factor from the usual dispersion curve becomes significant. It should be noted that there is no mention of combination frequencies in the paper by Sobel'man and one of the authors [3], nor in that by Faĭn and Khanin, who discuss the deformation of the amplification coefficient contour under the influence of a strong field. The appearance of combination frequencies in the polarization was first noted in [4] for the particular case of a two-level system with a single relaxation constant. Allusion to combination frequencies is also found in [2], where, however, the polarization at $\omega - \Omega$ is obtained to a higher order of perturbation theory than that at ω and $\omega + \Omega$. This is because Lamb considered all fields weak. Of course, in our calculation the combination frequencies are also lacking if the amplitude of the strong wave F becomes zero. Let us determine for what values of F the polarization at the combination frequency becomes comparable to that at the frequency $\omega + \Omega$. Clearly, this will happen when $|Fz_2| \approx |fX_2|$, that is, when

$$F^2 \geqslant \left| i\Omega + \tilde{\Gamma} + \frac{(\omega - \omega_0)^2}{i\Omega + \tilde{\Gamma}} \right| \left| i\Omega + \Gamma - \frac{\gamma^2}{i\Omega + \Gamma} \right|. \quad (15)$$

When $\Omega = 0$, condition (15) is identical with the condition that the field F result in saturation of the population difference [see (9)]. When Ω is large, a strong field F is required in order to satisfy (15).

Let us now consider the case of a non-monochromatic weak field

$$E = \frac{\hbar}{p} F \cos \omega t + \frac{\hbar}{p} \sum_j f^{(j)} \cos(\omega t + \Omega_j t + \varphi_j).$$

It is easy to verify that in the approximation assumed, i.e., linearity in the weak field, the term in the polarization of each component $f^{(j)}$ is independent of the remaining components, i.e., the polarization takes the form

$$p_{\text{av}} = p\,\mathrm{Re}\Big[2(X_3 + iX_4)e^{i\omega t} + \sum_j (z_3^{(j)} + iz_4^{(j)}) \exp\{i(\omega t + \Omega_j t + \varphi_j)\} + \sum_j (z_3^{(j)*} + iz_4^{(j)*}) \exp\{i(\omega t - \Omega_j t - \varphi_j)\}\Big]. \quad (16)$$

We call attention to the fact that the polarization $p_{\omega+\Omega}$ at some particular frequency $\omega + \Omega$ arises as a result of the effect upon the system of fields at both the frequencies $\omega + \Omega$ and $\omega - \Omega$. In this case it is essential that no other weak field components can influence $p_{\omega+\Omega}$. Therefore, a full description of the polarization at the frequency $\omega + \Omega$ for an arbitrary weak field can be obtained by considering two monochromatic weak fields at frequencies $\omega + \Omega$ and $\omega - \Omega$. Let us denote the amplitudes of these fields $f^{(1)}$ and $f^{(2)}$. Then, using (11), (12), (13), and (16), we find

$$p_{\omega+\Omega} = p \operatorname{Re}\left\{\left[i\frac{f^{(1)}X_2}{\tilde{\Gamma} + i(\omega + \Omega - \omega_0)} - iX_2 A\left(f^{(1)}\frac{\tilde{\Gamma} - i(\omega - \Omega - \omega_0)}{\tilde{\Gamma} - i(\omega - \omega_0)} + f^{(2)}\frac{\tilde{\Gamma} + i(\omega + \Omega - \omega_0)}{\tilde{\Gamma} + i(\omega - \omega_0)} e^{-i(\varphi_1+\varphi_2)}\right)\right] e^{i(\omega t + \Omega t + \varphi_1)}\right\},$$

$$A = \frac{F^2}{\tilde{\Gamma} + i(\omega + \Omega - \omega_0)} \frac{\tilde{\Gamma} + i\Omega/2}{\tilde{\Gamma} + i\Omega}\left\{\left[\tilde{\Gamma} + i\Omega + \frac{(\omega - \omega_0)^2}{\tilde{\Gamma} + i\Omega}\right]\left[\Gamma + i\Omega - \frac{\gamma^2}{\tilde{\Gamma} + i\Omega}\right] + F^2\right\}^{-1}. \quad (17)$$

It is apparent from (17) that the polarization depends on the phase relationships between the fields $f^{(1)}$ and $f^{(2)}$. If the frequency of the strong field and that of the transition are in resonance ($\omega = \omega_0$) the interference factor in (17) takes the form $f^{(1)} + f^{(2)} \exp\{-i(\varphi_1 + \varphi_2)\}$. If $\varphi_1 + \varphi_2 = \pi$ and $f^{(1)} = f^{(2)}$, this interference factor vanishes. In this case the polarization at the frequency $\omega + \Omega$ can be expressed as the product of the amplitude $f^{(1)}$, the population difference X_2, and the resonance factor $[\tilde{\Gamma} + i(\omega + \Omega - \omega_0)]^{-1}$. On the other hand, when $\varphi_1 + \varphi_2 = 0$ and $f^{(1)} = f^{(2)}$, the interference factor has its maximum modulus and the polarization is described by a function which differs considerably from a simple dispersion curve. In the general case the effect of a weak field of frequency $\omega - \Omega$ will depend upon $\varphi_1 + \varphi_2$, $f^{(2)}/f^{(1)}$, and the frequency of the strong field ω relative to that of the transition ω_0. Thus the manifestation of this effect can be quite different, depending upon the conditions of the particular physical problem.

In conclusion we should mention one possible application of these results. The development of quantum amplifiers has led to interest in the problem of the propagation of a modulated signal in the active medium. If the modulation depth is small, it is possible for the field to be strong at the carrier frequency and weak at the sideband frequencies. In this case the sideband frequencies are strongly coupled in phase to the carrier. Thus all the conditions described above can be realized in quantum amplifiers. In this case the previously-discussed mutual effect of weak fields comes into play. This signal-distortion mechanism should be taken into account in the theory of quantum amplifiers.

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Translated by L. C. Marquet
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