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Resonance processes in a two-level system in the presence of nonresonance fields

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A two-level system acted on by resonance and non-resonance fields is considered. It is shown that in a nonstationary regime the effect of nonresonance fields is proportional to the first power of the ratios of the amplitudes of the nonresonance fields to their detunings relative to resonance and depends on the initial phases of the fields. In a stationary regime in a system with damping the effect of nonresonance fields depends on the level of the resonance field. The analysis is based on the solution of the Bloch equations by the method of averaging, up to the third approximation of this method.

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There are well known resonance[1-6] and nonresonance[3-5,7] effects which appear when high-frequency fields interact with a quantum system. In the present paper we consider the behavior of a two-level system in fields which include simultaneously resonance and nonresonance frequencies. The main attention will be given to effects that arise when resonance and nonresonance fields act together. Effects of this kind occur when classical oscillators interact with high-frequency fields.[8,9]

The analysis of a two-level system in fields is based on the solution of the Bloch equations by the method of averaging, up to the third approximation of this method. It is well known[6] that the Bloch equations describe the behavior of a magnetized assembly of spins in the case of magnetic resonance. These equations are also used in optics[2] (the optical Bloch equations) in the study of the interaction of light with a two-level system. In optics one introduces an auxiliary vector of a fictitious electric spin $s=(s_1, s_2, s_3)$, or a speudospin vector whose components s_1 and s_2 are associated with the dipole moment of the system, while the third component s_3 is associated with inversion of the atom.[2]

1. Let us consider a two-level system described by the Bloch equations

$$\begin{aligned}\dot{s}_1&=-\omega_0 s_2-s_1/T_2,\\ \dot{s}_2&=\omega_0 s_1+\varkappa F(t)s_3-s_2/T_2,\\ \dot{s}_3&=-\varkappa F(t)s_2-(s_3-s_p)/T_1,\end{aligned}\tag{1}$$

where ω_0 is the frequency of the transition between the levels, $\varkappa$ is the gyromagnetic ratio in the case of magnetic resonance, and in optics $\varkappa=2d/\hbar$ (d is the magnitude of the dipole matrix element), $F(t)$ is the strength of the high-frequency magnetic (or in optics the electric) field acting on the system, T_1 is the longitudinal relaxation time (in optics the damping or inversion time), T_2 is the transverse relaxation time (in optics this is the damping time for the dipole moment), s_p is the equilibrium value to which the magnetization (the inversion) relaxes in the presence of noncoherent pumping in the case $F(t)\equiv 0$. A dot denotes differentiation with respect to the time. In the theory of magnetic resonance Eqs. (1) correspond to the case of orientation of the high-frequency magnetic field perpendicular to the static field. Here the components s_i are the projections of the magnetization vector.

Let the system be acted on by external linearly polarized fields

$$F(t)=F_1\cos(\omega_1 t+\zeta_1)+F_2\cos(\omega_2 t+\zeta_2),\tag{2}$$

where the field with frequency ω_1 is a resonance field, i.e., $\omega_1\approx\omega_0$, and that with frequency ω_2 is a nonresonance field, i.e., $\omega_2\neq\omega_0$. Substituting Eq. (2) in the equations (1) and using the substitution

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$$s_1 = u\cos(\omega_1 t+\zeta_1) - v\sin(\omega_1 t+\zeta_1),$$
$$s_2 = u\sin(\omega_1 t+\zeta_1) + v\cos(\omega_1 t+\zeta_1),$$
$$s_3 = w,$$

we arrive at a system of equations for $u, v,$ and ω that contains many-dimensional slow and fast motions; in the right hand sides of these equations a small parameter can be distinguished, owing to our assumptions that: 1) the frequency of inversion oscillations $\varkappa F_1/2$ in the rotating-wave approximation (the Rabi frequency) is small in comparison with the natural frequency ω_0, i.e., $\varepsilon = \varkappa F_1/(2\omega_0) \ll 1$, 2) the fractional detuning $\delta_0 = (\omega_1 - \omega_0)/\omega_0$ is small, 3) the dampings are small, i.e., $(\omega_0 T_1)^{-1}$, $(\omega_0 T_2)^{-1} \ll 1$.

The solution of this system can be found by using the method of averaging.[10] The solution consists of slowly varying functions $\bar{u}$, $\bar{v}, \bar{w}$ on which rapid oscillations are imposed. These vibrations are brought about by the existence of three nonresonance fields: A field with left circular polarization, which is part of the linearly polarized field with amplitude F_1, and two nonresonance circularly polarized fields which make up the field $F_2 \cos(\omega_2 t + \zeta_2)$. Besides oscillatory perturbations, the nonresonance fields produce cumulative effects on the system.

In the third approximation of the averaging method we find a set of equations for the averages $\bar{u}, \bar{v}, \bar{w}$:

$$\frac{d\bar{u}}{d\tau} = \delta\bar{v} - \frac{1}{\omega_0 T_2}\left[1 - \frac{\varepsilon^2}{2}\left(1 - \frac{T_2}{T_1}\right)Q\right]\bar{u},$$
$$\frac{d\bar{v}}{d\tau} = -\delta\bar{u} - \frac{1}{\omega_0 T_2}\left[1 - \frac{\varepsilon^2}{2}\left(1 - \frac{T_2}{T_1}\right)Q\right]\bar{v} + \varepsilon\bar{w}, \tag{3}$$
$$\frac{d\bar{w}}{d\tau} = -\varepsilon\left(1 - \frac{\varepsilon^2}{2}Q\right)\bar{v} - \frac{1}{\omega_0 T_1}\left[1 - \varepsilon^2\left(1 - \frac{T_1}{T_2}\right)Q\right]\bar{w} + \frac{1}{\omega_0 T_1}(1 - \varepsilon^2 Q)w_p,$$

where

$$\delta = \delta_0 - \frac{\varepsilon^2}{2}(P + \delta_0 Q),$$
$$P = \frac{1}{2\omega_1/\omega_0} + \frac{(F_2/F_1)^2}{(\omega_1+\omega_2)/\omega_0} + \frac{(F_2/F_1)^2}{(\omega_1-\omega_2)/\omega_0}, \tag{4}$$
$$Q = \frac{1}{4(\omega_1/\omega_0)^2} + \frac{(F_2/F_1)^2}{(\omega_1+\omega_2)^2/\omega_0^2} + \frac{(F_2/F_1)^2}{(\omega_1-\omega_2)^2/\omega_0^2}$$
$$\tau = \omega_0 t.$$

In the case $P = Q = 0$ we get from Eq. (3) the equations of the first approximation (the well known rotating-wave approximation), and with $Q = 0$ we get the equations of the second approximation. The original initial values u_0, v_0, w_0 undergo corrections; for the equations in the second approximation they can be written

$$\bar{u}(0) = u_0 + \varepsilon w_0 C, \quad \bar{v}(0) = v_0 - \varepsilon w_0 S,$$
$$\bar{w}(0) = w_0 + \varepsilon(v_0 S - u_0 C), \tag{5}$$

where

$$C = \frac{\cos 2\zeta_1}{2\omega_1/\omega_0} + \frac{F_2}{F_1}\frac{\cos(\zeta_1+\zeta_2)}{(\omega_1+\omega_2)/\omega_0} + \frac{F_2}{F_1}\frac{\cos(\zeta_1-\zeta_2)}{(\omega_1-\omega_2)/\omega_0},$$
$$S = \frac{\sin 2\zeta_1}{2\omega_1/\omega_0} + \frac{F_2}{F_1}\frac{\sin(\zeta_1+\zeta_2)}{(\omega_1+\omega_2)/\omega_0} + \frac{F_2}{F_1}\frac{\sin(\zeta_1-\zeta_2)}{(\omega_1-\omega_2)/\omega_0}. \tag{6}$$

The corrections on the initial conditions are due to the fact that in deriving Eq. (3) we impose on the functions that appear in the expansion of u, v, and w in powers of the small parameter the requirement that the averages of these functions over the rapidly varying phases must be zero.

In Eq. (4) the expression with the coefficient P is the well known Stark shift in the resonance detuning, which comes in in the second approximation of the averaging method. There are contributions to this shift both from the resonance field (Bloch-Siegert shift) and from the nonresonance field (quadratic dynamic Stark effect).

2. Let us examine the undamped case ($T_1^{-1} = T_2^{-1} = 0$) in the framework of the second approximation of the averaging method. Then the system of equations (3) gives a solution identical in structure with the well known Rabi solution in the rotating-wave approximation (cf., e.g., Ref. 2). Besides this shift in the resonance detuning, the nonresonance fields act through a specific mechanism which is expressed by the appearance in the expressions (5) of terms readjusting the initial values of u, v, w. Equations (5) and (6) show that the corrected initial values $u(0)$, $v(0)$, $w(0)$ give changes in the solution proportional to the first powers of the ratios of the nonresonance fields to their detunings relative to the resonance and depending on the initial phases ζ_1, ζ_2 of these fields. Without changing the oscillation frequencies of the functions u, v, w, this mechanism leads to a change of the amplitudes and initial phases of the functions u, v, w.

This mechanism can be observed in practice at radio frequencies in experiments on magnetic resonance. In optics the arbitrariness of the phases at the time the fields are turned on can be avoided, for example, for a beam of particles interacting with two fields from a single source, namely with the field incident on a reflector and the reflected field, which moves in the same direction as the particles. When one of the fields is at the resonance frequency the other is not, owing to the Doppler effect.

3. We shall analyze the stationary regime, on the basis of the third approximation of the averaging method. Stationary solutions which do not depend on the initial conditions follow from a system of algebraic equations which is obtained from Eq. (3) by setting $du/d\tau = dv/d\tau = dw/d\tau = 0$. These solutions are the well known expressions for u_r, v_r, w_r in the rotating-field approximation[2,6] (with the Stark effect taken into account) plus the corrections proportional to ε^2 which appear in the third approximation.

Let us compare the absorptions of $\bar{v}$ and v_r at their maxima. To do so we calculate, with $\delta = 0$ and to accuracy ε^2, the fractional difference

$$\frac{\bar{v} - v_r}{v_r} = -\frac{\varepsilon^2}{2}\frac{2T_1/T_2 + T_2/T_1 - 1 + (\varkappa F_1/2)^2 T_1 T_2}{1 + (\varkappa F_1/2)^2 T_1 T_2}Q, \tag{7}$$

which gives the decrease of the absorption in the stationary regime in the presence of the nonresonance fields [including the left-circular polarized field which is part of the field $F_1 \cos(\omega_1 t + \zeta_1)$] for arbitrary T_1,

T_2 and arbitrary level of the resonance field. The ratio (7) depends on the intensity of the resonance field, since the term $\varkappa F_1/2)^2 T_1 T_2$ in (7) can be of the order of or larger than unity. This fractional expression appears already in the rotating-field approximation,[2,6] where its increase with increasing field gives saturation.

Let us also compare the extremal values of $\bar{w}$ and w_r. Setting $\delta = 0$, we calculate to accuracy ε^2 the fractional difference

$$\frac{\bar{w}-w_r}{w_r}=-\varepsilon^2\frac{T_1/T_2+(\varkappa F_1/2)^2T_1T_2(1-T_2/2T_1)}{1+(\varkappa F_1/2)^2T_1T_2}Q, \tag{8}$$

which shows that the ratio depends on the level of the resonance field and that by changing the intensity of this field the sign of the ratio can be changed in the case $T_2 > 2T_1$. If we set $F_2 = 0$ in the expression for Q, the only contribution to the expression (8) is that from the left-circular polarized wave included in the resonance field $F_1 \cos(\omega_1 t + \zeta_1)$.

It follows from Eq. (3) that nonresonance fields change the widths of spectrum lines. For $\bar{v}$ and $\bar{w}$ the Lorentz curve is broadened in comparison with the case of a rotating resonance field only (we suppose that the intensity of the resonance field is the same in both cases) by a factor

$$1+\frac{\varepsilon^2}{2}\left[\frac{T_2/T_1-1+(\varkappa F_1/2)^2T_1T_2(T_2/2T_1-T_1/T_2)}{1+(\varkappa F_1/2)^2T_1T_2}Q-\frac{1}{4}\right]. \tag{9}$$

A characteristic feature is that the broadening caused by nonresonance fields depends on the level of the resonance field. Depending on the sizes of the relaxation times T_1, T_2 and the level of the resonance field, the nonresonance fields can lead to either a broadening or a narrowing of the spectral line. Setting $F_2 = 0$ in the expression for Q, we can obtain the contribution to the ratio (9) from only the left-circular wave which is one component of the field $F_1 \cos(\omega_1 t + \zeta_1)$.

In conclusion we note that the results derived both for the stationary and the nonstationary regimes by the method of averaging have been checked with exact calculations made by solving the initial equations (1) by the Runge-Kutta method on a computer.

There is no difficulty in extending the results to the case of many nonresonance fields.

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