Undamped nutation in a two-level system

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We consider the behavior of a two-level system acted upon by two resonant fields, one weak and the other strong. We obtain a generalization of Torrey's familiar solution for strong monochromatic resonant excitation. It is shown that the weak field produces in the system undamped nutation at the combination frequencies between the frequency difference of the initial fields and the harmonics of the Rabi frequencies. The harmonics of this nutation have amplitudes that depend resonantly on the amplitude and on the detuning of the strong field. Resonances set in when the Rabi-frequency harmonics are equal to the frequency difference of the initial fields. The results are generalized to include phase fluctuations of the weak field. They lead to a change of the undamped-nutation frequency. The positions of the nutation resonances and amplitudes remain unchanged, but their width depends on the width of the phase-fluctuation spectrum.

A general solution for the behavior of a two-level quantum system in a resonant external monochromatic field was obtained back in 1949 by Torrey.[1] This solution consists of a time-independent stationary part and a nonstationary one that describes transient damped processes, e.g., the oscillation of the difference between the level populations of the system (nutation). The damping is due to relaxation processes in the system. The stationary part of this solution was later variously generalized to include excitation by a bichromatic and, in general, a polychromatic intense field (see, e.g., Ref. 2 and the citations therein). It was found as a result, theoretically and experimentally, that if the modes of the external field are of equal intensity there are produced in the system quasilevels whose spacing is determined by combination relations between the intermode distance and the subharmonics of the Rabi frequency. These quasilevels cause the so-called subradiational structure in the absorption of the external field by the system.[3]

We obtain in the present system a solution, similar to Torrey's, for the density matrix elements of a system acted upon by two resonant fields, one weak and the other strong. The strong-field amplitude is such that nonlinear effects, such as saturation and the dynamic Stark effect,[4] are significant. The weak field leads in this case to the appearance of undamped nutation in the system. In addition, the subradiation spectrum in the absorption spectrum should be preserved also if the external-field mode intensities are not equal. It is proposed also that the weak-field phase fluctuates about its initial value. Such an approach permits, first, a more realistic description of the fields acting on the quantum system (for example, the emission of an intensity-stabilized single-mode laser is well described by a field model with a constant amplitude and with a random phase that follows the phase-diffusion model).[5] Second, it allows for effects not accounted for when the external fields are taken to be monochromatic. A monochromatic weak field, furthermore, is included in our field as a particular case when the width of the phase-fluctuation spectrum is zero.

Thus, the field interacting with the two-level system is represented in the form

$$\mathscr{E}(t)=\varepsilon_0\exp(i\omega t)+\zeta_0\exp i[\omega t+\Omega t+\varphi(t)+\varphi_0]+\text{c.c.},\qquad(1)$$

where ε_0 and ζ_0 are respectively the amplitudes of the strong and weak components. The strong-field frequency ω is close to the system transition frequency ω_0, i.e., $|\Delta| = |\omega-\omega_0| \ll \omega+\omega_0$. Furthermore, Ω is the difference between the field frequencies ($\Omega \ll \omega$) and $\varphi(t)$ is the random component of the weak-field phase, which shall describe using the phase-diffusion mode,[5] i.e., we assume that $\dot\varphi(t)=\mu(t)$ is a Gaussian δ-correlated random process:

$$\langle\mu(t)\rangle=0,\qquad\langle\mu(t_1)\mu(t_2)\rangle=2\gamma\delta(t_1-t_2),$$

$\langle\ \rangle$ denotes averaging over the random-process realizations, τ is the phase-fluctuation-spectrum matrix, and φ_0 is the initial phase of the weak field.

The dynamics of a two-level system in the field (1) is described in a rotating coordinate frame and in the approximation of slowly variable amplitudes by equations similar to Bloch's optical equations[6]:

$$\begin{aligned}
d\alpha(t)/dt&=-\alpha(t)/T_2-\Delta\beta(t)-a\zeta_0 n(t)\sin(\Omega t+\varphi(t)+\varphi_0),\\
d\beta(t)/dt&=-\beta(t)/T_2+\Delta\alpha(t)+a\varepsilon_0 n(t)+a\zeta_0 n(t)\cos(\Omega t+\varphi(t)+\varphi_0),\\
dn(t)/dt&=-(n(t)-n_0)/T_1-4a\varepsilon_0\beta(t)-4a\zeta_0[\alpha(t)\sin(\Omega t\\
&\quad+\varphi(t)+\varphi_0)+\beta(t)\cos(\Omega t+\varphi(t)+\varphi_0)],
\end{aligned}\qquad(2)$$

where

$$\alpha(t)=\operatorname{Re}\sigma_{21}(t),\quad\beta(t)=\operatorname{Im}\sigma_{21}(t),\quad n(t)=\sigma_{22}(t)-\sigma_{11}(t);$$

$\sigma_{ij}(t)$ are the slowly varying components of the system's density matrix, n_0 is population difference in the absence of external fields, T_2 and T_1 are the transverse and longitudinal relaxation times, $\Delta=\omega-\omega_0$ is the strong-field detuning, and a ε_0 and a ζ_0 are the interaction energies of the system with the corresponding fields in frequency units (interaction "frequencies"). For an electromagnetic field that leads to

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electrodipole transitions we have, e.g., $a = 2d/\hbar$, where d is the dipole matrix element.[6] For ultrasonic excitation of a quantum system, such as a paramagnetic impurity ion in a crystal, $a = GH_0/\hbar$, where G is the constant for the interaction between an external acoustic field and the effective spin of the ion (spin-phonon interaction) and H_0 is the intensity of the constant magnetic field that lifts the degeneracy of the magnetic-field energy levels.[7]

Regarding one field as weak compared with the other ($\zeta_0/\varepsilon_0 \ll 1$) we seek the solution of (2) in the form of two solutions, one in the absence of the weak field (at $\zeta_0 = 0$) and the other a correction to the first for the influence of the weak field $\zeta(t)$:

$$\alpha(t)=U(t)+u(t),\quad \beta(t)=V(t)+v(t),\quad n(t)=W(t)+w(t), \tag{3}$$

where $|u| \ll |U|$, $|v| \ll |V|$, $|w| \ll |W|$. In this case the system (2) breaks up into two sets of equations, for U, V, and W (zeroth approximation) and for u, v, and w (first approximation). Substituting (3) in (2) and retaining terms only of first order, we get

$$dX(t)/dt=KX(t)+L,\quad dx(t)/dt=Kx(t)+l(t), \tag{4}$$

where

$$X(t) = \begin{vmatrix} U(t) \\ V(t) \\ W(t) \end{vmatrix}, \quad x(t) = \begin{vmatrix} u(t) \\ v(t) \\ w(t) \end{vmatrix},$$

$$K = \begin{vmatrix} -T_2^{-1} & -\Delta & 0 \\ \Delta & -T_2^{-1} & a\varepsilon_0 \\ 0 & -4a\varepsilon_0 & -T_1^{-1} \end{vmatrix},$$

$$L = \frac{n_0}{T_1}\begin{vmatrix} 0 \\ 0 \\ 1 \end{vmatrix},$$

$$l(t) = a\zeta_0 \begin{vmatrix} W(t)\sin\Phi(t) \\ W(t)\cos\Phi(t) \\ -4[U(t)\sin\Phi(t) + V(t)\cos\Phi(t)] \end{vmatrix},$$

$$\Phi(t) \equiv \Omega t + \varphi(t) + \varphi_0.$$

The solutions (4) can be written in the form

$$X(t) = \exp(Kt)\left\{\int \exp(-Kt)\,L\,dt + \begin{vmatrix} 0 \\ 0 \\ n_0 \end{vmatrix}\right\}, \tag{5}$$

$$x(t)=\exp(Kt)\int \exp(-Kt)\,l(t)\,dt. \tag{6}$$

Since $\varphi(t)$ is a random function, the solutions (6) are also random functions. We are interested only in the mean values of these solutions:

$$\langle x(t)\rangle = e^{Kt}\int e^{-Kt}\langle l(t)\rangle dt. \tag{7}$$

Since $\varphi(t)$ obeys the phase-diffusion mode, we have $\langle e^{\pm i\varphi(t)}\rangle = e^{-\gamma t}$ (Ref. 8) and hence

$$\langle \sin\Phi(t)\rangle = e^{-\gamma t}\sin(\Omega t+\varphi_0),\quad \langle\cos\Phi(t)\rangle = e^{-\gamma t}\cos(\Omega t+\varphi_0). \tag{8}$$

Thus

$$\langle l(t)\rangle = a\zeta_0 e^{-\gamma t}\begin{vmatrix} W(t)\sin(\Omega t+\varphi_0) \\ W(t)\cos(\Omega t+\varphi_0) \\ -4[U(t)\sin(\Omega t+\varphi_0) + V(t)\cos(\Omega t+\varphi_0)] \end{vmatrix}. \tag{9}$$

To calculate $\exp(\pm Kt)$ in explicit form, we must find the eigenvalues of the matrices $\pm Kt$. The characteristic equation for the matrix Kt gives a cubic equation for the eigenvalues λ_i^+ ($i = 1,2,3$) of this matrix:

$$(\lambda^+)^3+(\lambda^+)^2\left(\frac{1}{T_1}+\frac{2}{T_2}\right)t+\lambda^+\left(\frac{1}{T_2^2}+\frac{2}{T_2T_1}+4a^2\varepsilon_0^2+\Delta^2\right)t^2 + \left(\frac{1}{T_1T_2^2}+\frac{4a^2\varepsilon_0^2}{T_2}+\frac{\Delta^2}{T_2}\right)t^3=0. \tag{10}$$

In the approximation in which the field $\varepsilon(t)$ is strong and the detuning is small (i.e., at $a\varepsilon_0 > T_2^{-1},\Delta$), and also at $T_2 \ll T_1$, the solutions of (10) are

$$\lambda_1^+=-\frac{t}{T_2}(1+\delta^2),\quad \lambda_2^+=-\frac{t}{2T_2}(1-\delta^2)+2ia\varepsilon_0(1+\delta^2)t,$$

$$\lambda_3^+=-\frac{t}{2T_2}(1-\delta^2)-2ia\varepsilon_0(1+\delta^2)t, \tag{11}$$

where $\delta^2 \equiv \Delta^2/8a^2/\varepsilon_0^2$. We obtain similarly the eigenvalues of the matrix $-Kt$:

$$\lambda_i^- = -\lambda_i^+. \tag{12}$$

The solution (5) is reduced with the aid of (11) and (12) to a form similar to the solution Torrey obtained[1] with the aid of Laplace transformation

$$X(t)=Ae^{-ct}+\left(B\cos st+\frac{C}{s}\sin st\right)e^{-bt}+D, \tag{13}$$

where

$$c=\frac{1}{T_2}(1+\delta^2),\quad b=\frac{1}{2T_2}(1-\delta^2),$$

$$s=(\Delta^2+4a^2\varepsilon_0^2)^{1/2}\approx 2a\varepsilon_0(1+\delta^2),$$

$$A = \frac{n_0}{4a\varepsilon_0}\begin{vmatrix} -\Delta(1-2\delta^2) \\ -(1+\delta^2T_1/T_2)/T_1 \\ (1+4\Delta^2T_2^2)/4a\varepsilon_0T_2^2 \end{vmatrix},$$

$$B = \frac{n_0}{4a\varepsilon_0}\begin{vmatrix} \Delta(1-2\delta^2) \\ \delta^2/T_2 \\ 4a\varepsilon_0(1-2\delta^2) \end{vmatrix},$$

$$\frac{C}{s} = \frac{n_0}{4a\varepsilon_0}\begin{vmatrix} -\Delta/4a\varepsilon_0T_2 \\ 8a\varepsilon_0(1-\delta^2) \\ T_2^{-1} \end{vmatrix},$$

$$D = \frac{n_0}{4a\varepsilon_0}\begin{vmatrix} -\Delta(1-3\delta^2)T_2/T_1 \\ (1+3\delta^2)/T_1 \\ (1+\Delta^2T_2^2)/a\varepsilon_0T_2T_1 \end{vmatrix}.$$

Solution (13) describes transient processes that attenuate as $t\to\infty$, and also the familiar components of the Bloch vector D in the stationary state, which describe in our approximation the saturation in the system.

A substantial deviation from (13) is obtained when the weak field $\zeta(t)$ is taken into consideration. The expression (6) is reduced with the aid of (11), (12), and (13) to the form

$$x(t)=n_0\frac{\zeta_0}{\varepsilon_0}\sum_{i=1}^{7}\sum_{j=1}^{7}\exp(-b_it)\,[B_{ij}\cos(s_jt+\varphi_0)$$
$$+C_{ij}\sin(s_jt+\varphi_0)\,], \qquad (14)$$

where

$$s_1=\Omega,\quad s_{2,3}=s\pm s_1,\quad s_{4,5}=2s\pm s_1,\quad s_{6,7}=3s\pm s_1; \qquad (15)$$

$$b_1=\gamma,\quad b_2=\frac{1}{T_2}(1+\delta^2),$$
$$b_3=\frac{1}{2T_2}(1-\delta^2)+\gamma,\quad b_4=-\frac{2\delta^2}{T_2}+\gamma,$$
$$b_5=-\frac{1}{2T_2}(1+3\delta^2)+\gamma, \qquad (16)$$
$$b_6=\frac{1}{2T_2}(1+3\delta^2)+\gamma,\quad b_7=\frac{1}{2T_2}(3+5\delta^2)+\gamma.$$

It follows from (14) that a weak field leads to the appearance of a large number of nutation frequencies (15) that connect by combination relations the frequency difference $s1$ of the initial fields with the harmonics of the Rabi system frequencies s. The damping also acquires qualitatively new properties. We investigate first the case when there are no phase fluctuations, i.e., we put $\gamma = 0$. It becomes necessary then to discard in (14) the terms with exponentials that have negative b_i, i.e., b_4 and b_5. These terms diverge with time. It can be seen from (14) and (16) that in the case of a monochromatic weak field the oscillations of the system's density-matrix elements have besides the damped harmonics also undamped ones (since $b_1 = 0$).

It can be seen from (16) that the phase fluctuations increase the damping coefficients b_1 and can, if the spectral width γ is large enough, eliminate completely the periodic factor $x(t)$. This calls for satisfaction of the inequality $b_i > s_i$.

Another important role of the phase fluctuations is the following. It can be seen from (16) that $b_4 = 0$ at $\gamma = 2\delta^2/T_2$ and $b_5 = 0$ at $\gamma = (1 + 3\delta^2)/2T_2$, i.e., undamped oscillations of the density-matrix elements set in again in the system at certain values of the phase-fluctuation spectrum. The physical meaning of this is simple—the damping increase due to the phase fluctuations compensates for the divergence of the exponentials $\exp(-b_{4,5}t)$. With further increase of γ the damping begins to prevail over the divergence, and these oscillations also become damped.

Expressions for the oscillation amplitudes B_{ij} and C_{ij} can be written in explicit form, but they are too unwieldy and will not be given here. A detailed investigation of the dependences of B_{ij} and C_{ij} on the phase-fluctuation spectrum width γ and on the detuning Δ leads to the following conclusions. First, the undamped oscillations in the absence of phase fluctuations (at $\gamma = 0$) occur at the frequencies s_1, s_4, and s_5, whereas for phase fluctuations with spectrum width $\gamma = (1 + 3\delta^2)/2T_2$ the frequencies of the undamped nutations are s_2 and s_3. The reason is that the corresponding coefficients in (14) ($i = 1$ and $i = 5$) are zero for the remaining harmonics. This fact can be used in principle to detect the presence of phase fluctuations in external fields. Second, it follows from (14) that the oscillation amplitudes are resonant. The resonances set in at $s_1 = 0$, $s = s_1$, $2s = s_1$, i.e., when the initial field frequencies are equal and when the difference of these frequencies coincides with harmonics of the frequency of the Rabi system s. As shown in Refs. 2 and 3, these resonances are manifestations of the splitting of the system's levels into quasilevels in an external field. To be sure, in these references the exciting-field mode intensities were assumed equal. Our calculations shown that a subradiation structure should appear also when one of the fields is weaker. Naturally, in this case the term (14) in the general solution (3) is small and bounded by the factor ζ_0/ε_0.

The locations of the aforementioned resonances are independent of the phase fluctuations, and only the widths of these resonances depend on the width of the fluctuation spectrum. In addition, resonances appear in the oscillation amplitudes also at $\Delta = 0$, at $\delta^2 = \gamma T_2 p1$, at $\delta^2 = 1 - 2\gamma T_2$, and at $\delta^2 = (2\gamma T_2 - 1)/3$, i.e., a resonant dependence on the spectrum width of the phase fluctuations sets in. This can also be used to determine the phase fluctuations of the field and their spectral widths.

To observe damped nutation in experiment it is necessary that the nutation period be shorter than the irreversible-relaxation times. Otherwise the presence of exponential relaxation damping leads to vanishing of the signal before even one nutation period takes place. No such restriction applies to observation of undamped nutation, and the frequencies (15) can be arbitrarily low. The strong-field amplitude, however, must be large enough for the resonant Rabi frequency a ε_0 to exceed the reciprocal transverse-relaxation time. This is necessary for the level splitting.[4] Since the dynamic Stark effect was successfully observed in optical fields,[9] this makes possible also observation of undamped nutation in this case. For ultrasonic excitation of a quantum system, for example, one can use an MgO crystal containing paramagnetic Ni^{2+} impurities. The interaction constant, the analog of the Rabi frequency, at a relative-strain field amplitude $\sim 10^{-7}$ at a frequency $10^{10}\ s^{-1}$ will be of the order of $10^7\ s^{-1}$, which also satisfies the condition $a\varepsilon_0 > T_2^{-1}$ (since $T_2 \sim 10^{-6}$ s, Ref. 7).

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