Optical transitions in a two-level system in the presence of a strong low-frequency field

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The emission and absorption of light by a two-level system in the presence of a strong electromagnetic field of frequency much smaller than the eigenfrequency of the system is investigated by using the adiabatic approximation. It is demonstrated that the dependence of the probabilities for these processes on the intensity of the strong field differs from that predicted by perturbation theory. The limits of validity of the latter are established.

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Enlistment of the concept of quasienergy, introduced by Zel'dovich[1] and Ritus,[2] produces an especially intuitive picture of optical transitions in quantum systems in the presence of a strong electromagnetic field, considered as a periodic perturbation. The fruitfulness of the quasienergy approach, which utilizes only symmetry considerations, is due in particular to the possibility of quantitatively analyzing the processes which occur, without making assumptions about the magnitude of the interaction of the system with the variable perturbation. However, a quantitative calculation of the optical transitions in the quasienergy spectra of bound systems runs into serious difficulties, especially for processes involving the participation of distant satellites of the quasienergy (a large number of photons of the strong field), when the use of perturbation theory in the magnitude of the interaction is inconvenient, and its criteria for strong fields are either unclear or are certainly violated. A number of articles[3-10] are devoted to the calculation of multiphoton transitions in bound systems without using perturbation theory; of these the work of Zaretskiĭ and Kraĭnov[10] is closest to the work expounded below.

In the present article the emission and absorption of light by a two-level system (the separation between the levels is ω_0) are investigated in a strong electromagnetic field of frequency ω, where it is assumed that $\omega_0/\omega \equiv \Delta \gg 1$. In this connection the emission of light by the system may also be treated as multiphoton Raman scattering of the field ω. The energy levels of the system (1 denotes the lower level and 2 is the upper level) are assumed to be nondegenerate and coupled by a dipole interaction. A qualitative picture of the emission of light by such a system may be obtained from the following analogy with the well known problem in radio engineering concerning the emission of a frequency modulated signal. In a constant field the energy levels of the system are repelled, and the system radiates at the frequency $\nu = \omega_0 + \delta\omega_0$. If the external field slowly changes, according to a harmonic law for example, the increase $\delta\omega_0$ of the system's eigenfrequency becomes a periodic function of the time. If this frequency-modulated emission of the system is passed through a spectral analyzer, a central frequency $\nu_0 = \overline{\nu(t)} = \omega_0 + \overline{\delta\omega_0(t)}$ (the bar denotes averaging over the period of modulation) and the harmonics $\nu_k = \nu_0 \pm k\omega$ (k takes integer values) are observed in the spectrum of the radiation. Moreover, the relation between the intensity of the fundamental frequency ν_0 and the intensities of the harmonics ν_k is determined by the so-called depth of the modulation. For a sufficiently large depth of modulation, the intensities of certain harmonics exceed the intensity of the fundamental frequency. The fundamental frequency is suppressed.

Let us quantitatively investigate the process of the absorption and emission of light by the system considered above. The natural separation of fast and slow subsystems for $\Delta \gg 1$ makes utilization of the adiabatic approximation appropriate. The adiabatic wave functions have the form (see, for example,[11, 12])

$$\Psi_1 = \left(|1\rangle \cos\frac{\beta(t)}{2} + |2\rangle \sin\frac{\beta(t)}{2}\right)\exp\left\{i\int\Omega(t)\,dt\right\}$$
$$\Psi_2 = \left(-|1\rangle \sin\frac{\beta(t)}{2} + |2\rangle \cos\frac{\beta(t)}{2}\right)\exp\left\{-i\int\Omega(t)\,dt\right\}; \qquad (1)$$
$$\Omega(t) = {}^1/_2\omega_0\sqrt{1+q^2\sin^2\omega t};$$
$$\mathrm{tg}\,\beta(t) = q\sin\omega t; \quad q = 2e_0F_0Z_{12}/\omega_0;$$

$|1\rangle$ and $|2\rangle$ denote the eigenstates of the system in the absence of external fields, corresponding to the energies $\mp\omega_0/2$; F_0 denotes the intensity of the field ω, for which Z-polarization is assumed[1)] (here and below we assume $\hbar = 1$).

Each of the functions (1) may be represented in the form

$$\Psi_{1,2} = \exp\{-iE_{1,2}t\}\chi_{1,2}(t), \quad \chi_{1,2}(t) = \chi_{1,2}(t+2\pi/\omega),$$

$$E_{1,2} = \mp\frac{1}{2}\omega S, \quad S = \frac{\Delta}{\pi}\int_0^\pi (1+q^2\sin^2 x)^{1/2}\,dx,$$

$E_{1,2}$ denote the quasienergies of the system, obtained in the adiabatic approximation. Expressing the interaction of the system with a weak optical field of frequency ν in the form Q sin νt, we find the following result for the probability of absorption per unit time of the light ν in the transition $1 \to 2$:

$$W_{12}(\nu) = \frac{\pi|Q_{12}|^2}{2\omega}|U|^2\sum_n \delta(S-p-n), \quad p = \frac{\nu}{\omega} \qquad (2)$$

$$U = \frac{1}{2\pi i}\int_C \frac{dz}{(1-q^2\,\mathrm{sh}^2 z)^{1/2}}\exp\left\{pz - \Delta\int_0^z (1-q^2\,\mathrm{sh}^2 z')^{1/2}\,dz'\right\}. \qquad (3)$$

The integration contour in (3) consists of the segments $(\infty - i\pi, -i\pi)$, $(-i\pi, i\pi)$, and $(i\pi, i\pi, +\infty)$. The contour integral (3) can by evaluated by the method of steepest descents. However, it is not possible to obtain a single analytic expression which is valid for arbitrary relationships between the parameters p, Δ, and q; therefore, we consider the following possible situations separately (later $r = |p^2 - \Delta^2|^{1/2}$ everywhere).

1a. $p > \Delta$, $r/q\Delta > 1$. Here the saddle points are determined by the formula

$$z_0 = -\mathrm{Arch}\,(r/q\Delta) \pm i\pi/2. \qquad (4)$$

The directions of quickest descent from the saddle points make an angle $\pi/2$ with the real axis. Evaluation of the integral (3) with formula (4) taken into consideration, after the appropriate deformation of the integration contour, gives

$$W_{12}(\nu)=\frac{|Q_{12}|^2\Delta^2}{\omega p y_1}\Phi^2\left(\frac{\pi}{2}\sqrt{\frac{y_1}{2p}}\right)\exp\{g_1(q,\Delta,p)\}\sum_n\cos^2\frac{\pi n}{2}\delta(S-p-n),$$

$$y_1=\sqrt{(p^2-\Delta^2)[p^2-\Delta^2(1+q^2)]},\tag{5}$$

$$g_1(q,\Delta,p)=\frac{2\Delta}{(1+q^2)^{1/2}}D\left(\varepsilon,\frac{1}{(1+q^2)^{1/2}}\right)-2p\,\mathrm{Arch}\frac{r}{q\Delta}+\frac{2py_1}{r^2},$$

$$k^2D(\varepsilon,k)=F(\varepsilon,k)-E(\varepsilon,k),\quad \varepsilon=\arccos(\Delta q/r),\tag{6}$$

$F(\epsilon, k)$ and $E(\epsilon, k)$ are elliptic integrals of the first and second kinds, and $\Phi(x)$ is the probability integral. The factor $\cos^2(\pi n/2)$, expressing the selection rule (n is even), appears due to the interference of the contributions of the saddle points.

1b. $p > \Delta$, $r/q\Delta < 1$, $q \ll 1$. The saddle points are given by

$$z_0=\pm i\rho,\ i\rho-i\pi,\ -i\rho+i\pi;\ \rho=\arcsin(r/q\Delta).\tag{7}$$

The directions of quickest descent make angles $\pm\pi/4$ with the real axis. Integration gives

$$W_{12}(\nu)=\frac{|Q_{12}|^2\Delta^2}{\omega p y_2}\cos^2\left[g_2(q,\Delta,p)-\frac{\pi}{4}\right]\left\{\Phi\left[\left(\frac{\pi}{2}-\rho\right)\left(\frac{y_2}{p}\right)^{1/2}\right]+\Phi\left(\rho\left(\frac{y_2}{p}\right)^{1/2}\right)\right\}^2\sum_n\cos^4\frac{n\pi}{2}\delta(S-p-n),\tag{8}$$

$$y_2=\sqrt{(p^2-\Delta^2)[\Delta^2(1+q^2)-p^2]},$$

$$g_2(q,\Delta,p)=p\rho-\Delta(1+q^2)^{1/2}\left[E\left(\frac{\pi}{2},\frac{q}{(1+q^2)^{1/2}}\right)-E\left(\gamma,\frac{q}{(1+q^2)^{1/2}}\right)\right],\tag{9}$$

$$\gamma=\pi/2-\rho.$$

2a. $p < \Delta$. The saddle points are given by

$$z_0=\mathrm{Arsh}\,(r/q\Delta)+ik\pi,\quad k=0,\ \pm1.\tag{10}$$

Here one must keep in mind the possibility of incidence of the branch points of the expression $\psi(z) = (1-q^2\sinh^2 z)^{-1/2}$ given by $z_0' = \sinh^{-1}(1/q) + ik\pi$, $k = 0, \pm 1, \ldots$, into the "regions of influence" (see[13]) of the saddle points z_0.

One can verify that for $q < 1$ and

$$\ln(\Delta/r)>(2p)^{1/2}/r\tag{11}$$

(a rough estimate of p from (11) gives $p > 2\Delta^{2/3}(e-1)^{2/3}$) the points z_0' turn out to be outside the "regions of influence" of the saddle points z_0 and one can take $\psi(z)$ outside the integration sign at the saddle points. In this case the directions of steepest descent make an angle of $\pi/2$ with the real axis. We find

$$W_{12}(\nu)=\frac{|Q_{12}|^2\Delta^2}{\omega p y_3}\Phi^2\left(\frac{\pi}{2}\left(\frac{y_3}{2p}\right)\right)^{1/2}\exp\{g_3(q,\Delta,p)\}\sum_n\cos^4\frac{n\pi}{2}\delta(S-p-n),$$

$$y_3=\sqrt{(\Delta^2-p^2)[\Delta^2(1+q^2)-p^2]},\tag{12}$$

$$g_3(q,\Delta,p)=2p\,\mathrm{Arsh}\frac{r}{q\Delta}-\frac{2\Delta}{(1+q^2)^{1/2}}D\left(\delta,\frac{1}{(1+q^2)^{1/2}}\right)-\frac{2pr^2}{y_3},$$

$$\delta=\arcsin\left(\frac{1+q^2}{1+\Delta^2q^2/r^2}\right)^{1/2}.\tag{13}$$

2b. For a sufficiently small ratio p/Δ, condition (11) is violated and the points z_0' fall inside the "regions of influence" of the points z_0. In this case one can, without going outside the limits of the "regions of influence", deform the integration contour in such a way that it passes through the points z_0', going around them along arcs of small radius. In this case the directions of quickest descent make angles $\pm\pi/3$ with the real axis. Integration with these characteristics taken into consideration gives

$$W_{12}(\nu)=A\frac{3^{2/3}|Q_{12}|^2}{8\pi\omega[\Delta(1+q^2)]^{2/3}}\exp\{g_3(q,\Delta,p)\}\sum_n\cos^4\frac{n\pi}{2}\delta(S-p-n),\tag{14}$$

$$A=\left[\sum_{n=0}^{\infty}\frac{1}{n!(3n+1)}\right]^2.\tag{15}$$

The series determining A converges rapidly, $A \approx 1.7$.

The entire range of variation of the frequency $\nu = \omega p$ of absorbable light is exhausted by case 2b.

We find the following result for the probability of multiphoton Raman scattering of a low-frequency field ω by the system in state $2-W_{21}^e$ (the emission of light by the system, accompanied by the transition $2 \to 1$, in the presence of the low-frequency field ω):

$$W_{21}^e=\sum_{n=-\infty}^{n_0}W_{21}^e(n),\quad n_0=[\Delta],\tag{16}$$

$$W_{21}^e(n)=4e_0^2\omega^3(S-n)^3c^{-3}|Z_{21}|^2|U_n|^2,$$

$$U_n=U|_{p=S-n},$$

$[\Delta]$ denotes the integer part of the number Δ. The quantity U is defined by formula (3) and may be obtained, respectively, from Eqs. (5), (8), (12), and (14) in each of the situations considered above. As follows from (16), the harmonics of the fundamental frequency ω_0 are present in the spectrum for the Raman scattering of the low-frequency radiation ω by the system, where both the frequencies of the harmonics and their intensities are complicated functions of the intensity F_0 of the field ω. Formulas can be derived in analogous fashion for the Raman scattering of radiation ω by the system in its ground state 1 (the emission of light by the system in its ground state in the presence of the low-frequency field ω, accompanied by the transition $1 \to 2$). Such a process, however, requires the participation of at least $n_0 + 1$ photons of frequency ω and has a considerably smaller probability than that described by formula (16).

Let us consider certain limiting cases. Let $q \ll 1$ and $p < \Delta$. In particular, from formula (12) we obtain the following result to the first nonvanishing order of the expansion in powers of q:

$$W_{12}(\nu)=\frac{|Q_{12}|^2\Delta^2}{\omega p(\Delta^2-p^2)}\left(\frac{\Delta+p}{2\Delta}\right)^{2p}\left(\frac{e^2q^2}{16}\frac{\Delta+p}{\Delta-p}\right)^{\Delta-p}\sum_n\cos^4\frac{n\pi}{2}\delta(\Delta-p-n).\tag{17}$$

(In the derivation of expression (17), it was additionally assumed that $n \geq 1$.) According to Eq. (17), $W_{12}(\nu) \sim F_0^{2n}$, as should happen upon using ordinary n-th order perturbation theory for a description of a process involving the participation of n quanta of the field ω. Actually formula (17) is applicable under the restriction $\Delta q^2 \ll 1$, which is much more stringent than $q \ll 1$. For $\Delta q^2 > 1$, as one can easily verify, a deviation from the power law $W_{12}(\nu) \sim F_0^{2n}$ occurs toward the side of a weakening of the dependence of the transition probability on the field intensity F_0 ($W_{12}(\nu) \sim F_0^{2(n-n')}$, where $n' \equiv n'(F_0) > 0$). For sufficiently large values of the parameter Δq^2, it is necessary to use the general formula (12) directly.

Let us analyze formula (8) upon fulfillment of the conditions $q \ll 1$, $\Delta q^2 > 1$. Let $n = 0$ (the transition goes without real absorption or emission of quanta ω). We find

$$W_{12(n=0)}(\nu)=\frac{8|Q_{12}|^2}{\omega pq^2}\cos^2\left(\frac{\Delta q^2}{8}-\frac{\pi}{4}\right)\delta\left(p-\Delta-\frac{\Delta q^2}{4}\right).\tag{18}$$

According to Eq. (18) the probability of absorbing the light ν at the fundamental frequency ($\nu = \omega_0 + \omega_0 q^2/4$) is a pulsing function of the intensity F_0 of the field ω

($\omega_0 q^2/4$ obviously represents the dynamical Stark shift of the system's eigenfrequency). This result (for n = 0) was previously obtained by Kovarskiĭ[7] by another method and is called the effect of suppression. (For comparison, informula (30) of his article[7] it is necessary to use the asymptotic representation of the Bessel function $J_0(x) \approx (\sqrt{2/\pi x})^{1/2} \cos(x - \pi/4)$ for $x > 1$). In the more general case $n \neq 0$ the pulsed dependence of $W_{12}(\nu)$ on F_0 for $\Delta q\ r > 1$ is expressed by formula (8).[2)] Analysis of formulas (5) and (14) also demonstrates a deviation from the power-law dependence, predicted by ordinary perturbation theory, of the transition probability on the intensity F_0 of the field ω with increasing F_0.

3. Let us consider the transition $1 \rightarrow 2$ under the influence of a very strong field ω (without the participation of photons ν). In lowest order adiabatic perturbation theory the probability of such a process is determined by the formula

$$W_{12}(\omega)=\frac{1}{2}\pi\omega q^2|U|^2\sum_n \delta(S-n), \tag{19}$$

$$U=\frac{1}{2\pi i}\int_c \frac{dz\,\mathrm{ch}\,z}{1-q^2\,\mathrm{sh}^2 z}\exp\left\{-\Delta\int_0^z (1-q^2\,\mathrm{sh}^2 z')^{1/2}\,dz'\right\}. \tag{20}$$

The integral (20) is evaluated by the saddle-point method. The saddle points $\tilde{z}_0 = \sinh^{-1}(1/q) + ik\pi$, $k = 0, \pm 1$, coincide with the poles of the integrand in (20). In order to take this property into account, the integration contour is chosen in the same way as in case 2b. We find

$$W_{12}(\omega)=\left(\frac{\pi}{3}\right)^2\frac{2\omega}{\pi}\exp\left\{-\frac{2\Delta}{(1+q^2)^{1/2}}D\left(\frac{\pi}{2},\frac{1}{(1+q^2)^{1/2}}\right)\right\} \times\sum_n \sin^4\frac{n\pi}{2}\,\delta(S-n). \tag{21}$$

The use of lowest order adiabatic perturbation theory in order to obtain the transition probability in a two-level system under the influence of a slowly varying perturbation is not a completely rigorous procedure. As is well known (see, for example,[11, 14]) the contributions to the transition probability coming from the higher-order terms in adiabatic perturbation theory turn out to be of the same order as the contribution of the lowest-order term (more correctly, only numerically smaller than the contribution of the lowest order term). This is related to the fact that the adiabatic functions (1) poorly describe the system at the points of complex time, corresponding to branch points of the expression $\Omega(t)$. At the same time, in case (3) it is precisely these points which substantially determine the transition probability,[10] which leads to the necessity of summing the entire series of perturbation theory with respect to non-adiabaticity, which is equivalent to use of the formula

$$W \sim \exp\left\{-2\,\mathrm{Im}\int_0^{\tilde{z}_0} \Delta(1-q^2\,\mathrm{sh}^2 z)^{1/2}dz\right\}$$

(see[11, 15]). This formula allowed Zaretskiĭ and Kraĭnov[10] to obtain for case (3) an expression of the form (21) with the correct preexponential factor–unity, whereas the first nonvanishing order of adiabatic perturbation theory (formulas (19) and (20)) gives an inaccurate preexponential factor, $(\pi/3)^2 \approx 1.1$.

Everything that has been said also pertains to case 2b, when the saddle points are close to the branch points of the quantity $\Omega(t)$. In accordance with this, formula (14) contains the correct exponential factor; however, the preexponential factor in Eq. (14) is inaccurate. At the same time, as follows from a comparison with case 3, it evidently should be close to the true value.

A different situation arises in cases 1a, 1b, and 2a. Here the saddle points do not coincide with the branch points of $\Omega(t)$ and therefore lie in regions of complex time where the adiabatic functions (1) describe the system well. This property is more strongly expressed, the larger the adiabatic parameter Δ is. This gives the reason to assume that the formulas derived in cases 1a, 1b, and 2a possess greater preexponential accuracy than in cases 2b or 3, the accuracy being greater the larger the value of Δ.

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1) In order to not go outside the framework of the adiabatic approximation, we shall assume below that the intensity F_0 is much less than atomic field strengths, i.e., $q \ll 1$ (see [10]).

2) We note that in this case the parameter $\Delta q^2/8$ is obviously analogous to the depth of the modulation in the radio engineering problem concerning the emission of a frequency-modulated signal.

1. Ya. B. Zel'dovich, Zh. Eksp. Teor. Fiz. 51, 1492 (1966) [Sov. Phys.–JETP **24**, 1006 (1967)]; Usp. Fiz. Nauk 110, 139 (1973) [Sov. Phys.-Uspekhi 16, 427 (1973)].
2. V. I. Ritus, Zh. Eksp. Teor. Fiz. 51, 1544 (1966) [Sov. Phys.-JETP **24**, 1041 (1967)].
3. L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys.-JETP **20**, 1307 (1965)].
4. A. M. Perelomov, V. S. Popov, and M. V. Terent'ev, Zh. Eksp. Teor. Fiz. 50, 1393 (1966) [Sov. Phys.-JETP **23**, 924 (1966)]; Zh. Eksp. Teor. Fiz. 51, 309 (1966) [Sov. Phys.-JETP **24**, 207 (1967)].
5. A. I. Nikishov and V. I. Ritus, Zh. Eksp. Teor. Fiz. 50, 255 (1966) [Sov. Phys.-JETP **23**, 168 (1966)]; Zh. Eksp. Teor. Fiz. 52, 223 (1967) [Sov. Phys.-JETP **25**, 145 (1967)].
6. V. A. Kovarskiĭ, Zh. Eksp. Teor. Fiz. 57, 1217 (1969) [Sov. Phys.-JETP **30**, 663 (1970)].
7. V. A. Kovarskiĭ, Zh. Eksp. Teor. Fiz. 57, 1613 (1969) [Sov. Phys.-JETP **30**, 872 (1970)].
8. Yu. A. Bychkov and A. M. Dykhne, Zh. Eksp. Teor. Fiz. **58**, 1734 (1970) [Sov. Phys.-JETP 31, 928 (1970)].
9. V. A. Kovarskiĭ and N. F. Perel'man, Zh. Eksp. Teor. Fiz. **60**, 509 (1971) [Sov. Phys.-JETP 33, 274 (1971)].
10. D. F. Zaretskiĭ and V. P. Kraĭnov, Zh. Eksp. Teor. Fiz. **66**, 537 (1974) [Sov. Phys.-JETP **39**, 257 (1975)].
11. A. M. Dykhne, Zh. Eksp. Teor. Fiz. **41**, 1324 (1961) [Sov. Phys.-JETP **14**, 941 (1962)].
12. E. E. Nikitin, Teoriya elementarnykh atomno-molekulyarnykh protsessov v gazakh (Theory of Elementary Atomic and Molecular Processes in Gases), "khimiya", 1969, p. 108 (English Transl., Oxford Univ. Press, 1974).
13. N. G. De Bruijn, Bibliotheca Mathematica: Asymptotic Methods in Analysis, Vol. 4, American Elsevier Publishing Co., Inc., 1958 (Russ. Transl., IIL, M. 1961).
14. A. M. Dykhne, Zh. Eksp. Teor. Fiz. **38**, 570 (1960) [Sov. Phys.-JETP **11**, 411 (1960)].
15. V. L. Pokrovskiĭ, S. K. Savvinykh, and F. R. Ulinich, Zh. Eksp. Teor. Fiz. **34**, 1272 (1958)[Sov. Phys.-JETP **7**, 879 (1958)].

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