# Nonadiabatic transition in a broad-spectrum radiation field

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We investigate the excitation, by nonmonochromatic radiation, of a two-level system whose transition frequency varies linearly with time in the vicinity of the spectral band of the field. It is shown that the character of certain nonlinear optical phenomena in systems of colliding atoms depends substantially on the nonmonochromaticity factor. In particular, the phenomenon of "bleaching of a medium without saturation" can be observed only if the excitation is by a sufficiently narrow line, whereas broadband pumping leads to saturation of the atomic system.

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1. The study of optical phenomena in systems of colliding atoms (excimers, quasimolecules) is of interest in various branches of physics. The theory of processes that are linear in the field intensity was developed for the most part in the early Fifties. Recently, nonlinear optical phenomena in these systems have been attracting interest. A constructive theory of these phenomena[1] was developed in the adiabatic approximation. It is assumed that the position of the resonant atomic line (of the frequency of the optical transition) is determined by the distance between atoms that move along classical trajectories. Optical excitation of colliding atoms can be described in this approximation within the framework of the model of a two-level system with time-varying transition frequency. If the system is excited by monochromatic radiation that is quasi-resonant with the frequency of the atomic transition, the use of the rotating-field approximation reduces the problem of describing the optical excitation to the problems of the asymptotic theory of atomic collisions.[2,3]

Greatest interest attaches to nonlinear phenomena in radiation fields whose frequency falls in the region of the static wing of the atomic line. In this case the effective action of the field on the atoms takes place at collisions such that the frequency of the atomic transition passes through the vicinity of the excitation frequency. The probability of exciting the atoms on passing through the "resonance" region (the nonadiabaticity region) is described by the Zener formula.[2] The two passages through the nonadiabaticity region as the atoms approach each other and then move apart cause the interesting phenomenon[1] of "bleaching the medium without saturation." The gist of this phenomenon is that the excitation of the atomic medium decreases with increasing intensity of the atomic medium as the intensity of the effective field increases, starting with a certain critical value.

In the description of the process of optical excitation of quasimolecules it is customary to neglect the influence of the relaxation of the atomic states on the transition probability, which is determined by the Landau–Zener–Stückelberg theory. Of course, relaxation processes of the radiative type are of no importance in these systems, for in fact they cannot evolve during the time of passage through the nonadiabaticity region ($\approx 10^{-12}$ sec for the systems discussed in the literature[1,4]). In optical excitation of an atomic transition by the radiation from real intense sources, however, account must be taken of the monochromaticity factor, whose role is analogous, to a certain degree, to that of relaxation.[5] The influence of relaxation on the character of a nonadiabatic transition in a two-level system was recently considered in Refs. 6 and 7. Unfortunately, the reduction of the action of the monochromaticity factor to the action of relaxation is in the general case not a trivial matter, and is all the more complicated in the case of systems whose transition frequency varies with time.

In the present study we investigated theoretically and experimentally the excitation, by an intense nonmonochromatic field, the radiation of a two-level system whose transition frequency varies linearly with time in the vicinity of the spectral band of the exciting radiation. The results differ substantially from those of the theory of monochromatic excitation. In particular, it is shown that the phenomenon of "bleaching without saturation" is realized upon satisfaction of a condition that limits the widths of the acting radiation.

2. We investigate first the situation theoretically. The dynamics of a two-level system in a nonmonochromatic field

$$\mathscr{E}(t)=E(t)e^{i\omega_0 t}+\text{c.c.} \tag{1}$$

is described in the resonance approximation by an equation for the density matrix in the form

$$\dot{n}=4\mathrm{Re}\,(iF^*f_{12}),\quad n=f_{11}-f_{22};$$
$$\dot{f}_{12}=i\omega(t)f_{12}+iFn. \tag{2}$$

Here $\omega(t)$ is the instantaneous shift of the atomic transition frequency $\omega_{21}$ relative to $\omega_0$: $\omega(t)=\omega_{21}(t)-\omega_0$, and $F(t)=dE/\hbar$, where the dipole matrix element $d$ of the transition is assumed to be independent of the interatomic distance and of the time; $E(t)$ is a stationary

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random function of $t$.

Eliminating from the first equation of the system (2) the off-diagonal matrix elements, provided that the field is turned-on adiabatically and there is no coherence at $t = -\infty$, we write down the equation for the population difference in the form

$$\dot{n}(t) = -4\,\mathrm{Re}\int_{-\infty}^{t} F(t)F^*(t')n(t')\exp\left[i\int_{t'}^{t}\omega(t_1)\,dt_1\right]dt'. \tag{3}$$

Equation (3) contains random functions of the time, and therefore $n(t)$ is also a random function. Physical interest attaches to the quantity $\langle n\rangle$ averaged over the ensemble of the realizations of the random field. An equation in closed form can be obtained for $\langle n\rangle$ by assuming that the random field varies so rapidly that the population is unable to follow these rapid changes. In this case the changes of $n(t)$ and $F(t)$ can be regarded as uncorrelated[1], and to obtain an equation for $\langle n\rangle$ from (3) we can use the correlation-decoupling approximation

$$\langle\dot{n}(t)\rangle = -4\,\mathrm{Re}\int_{-\infty}^{t} a^2 r(t-t')\langle n(t')\rangle\exp\left[i\int_{t'}^{t}\omega(t_1)\,dt_1\right]dt', \tag{4}$$

where $a^2 r(t-t') = \langle F(t)F^*(t')\rangle$ is the correlation function of the field and is connected with the spectrum of the effective field by a Fourier transformation; $a^2$ is a quantity proportional to the average value of the effective radiation, and $r(0) = 1$. We shall assume henceforth that $r(t)$ is characterized by a single time scale $\tau_c$ (the correlation time) and that at $|t| > \tau_c$ the function $r(t)$ decreases rapidly.

With an aim at an eventual comparison with the result of the Landau–Zener theory, we assume

$$\omega(t) = bt, \tag{5}$$

so that the resonance between the transition and the field is reached in the vicinity of $t = 0$.

To keep the result of the solution of (4) from exceeding the bounds of the correlation-decoupling approximation, it suffices to confine oneself to the case of a small change of population in the scale of $\tau_c$. This is precisely the case considered below. We note incidentally that Eq. (4), in the opposite limiting case, as $\tau_c \to \infty$ and at $r(t) = 0$, becomes identical with the equation that describes the transition in a monochromatic field. The fact that the equations become identical, however, does not justify the assumption that Eq. (4) describes correctly even qualitatively the transition in a quasistatic field.[2]

In view of our assumption that $\langle n\rangle$ changes slowly in the scale of $\tau_c$ and that $r(t)$ decreases rapidly with time, we take $\langle n(t')\rangle$ in Eq. (4) outside the integral sign at the point $t' = t$. The solution of the equation obtained in this manner (with the substitution $t - t' = s$)

$$\langle\dot{n}(t)\rangle = -4a^2\langle n(t)\rangle\int_0^\infty ds\, r(s)\cos(\tfrac{1}{2}bs^2 - bst) \tag{6}$$

subject to the initial condition $n(-\infty) = 1$, i.e., in the case when the first term is fully populated, determines the quantity

$$\langle n(\infty)\rangle = e^{-2\delta}, \tag{7}$$

where $\delta = 2\pi a^2/b$. The population of the second term, after a single passage through the resonance point (the transition probability) is

$$w_w = \tfrac{1}{2}(1 - e^{-2\delta}). \tag{8}$$

Let us determine the region of applicability of the result. Estimating the right-hand of (6) at

$$|\langle\dot{n}\rangle/\langle n\rangle| < 4a^2\int_0^\infty r(s)\,ds = 4a^2\tau_c, \tag{9}$$

we obtain the sufficient condition

$$4a^2\tau_c^2 \ll 1, \tag{10}$$

which limits the permissible values of the intensity at a given width of the spectrum of the effective field.

3. The experimental investigation of the processes considered above in the optical band encounters a number of major difficulties, due mainly to the short time of passage of the colliding atoms through the nonadiabatic region ($\approx 10^{-12}$ sec). For a detailed comparison with the result (7) and (8) we have performed experiments that simulate the excitation of the colliding atoms by a nonmonochromatic radiation field.

The model research object was taken to be oriented vapor of atomic $^{113}$Cd. This isotope of Cd has in the ground state an angular momentum $I = \frac{1}{2}$. In a constant magnetic field $H = 5$ G the frequency of the Zeeman splitting between the magnetic sublevels $|+\frac{1}{2}\rangle$ and $|-\frac{1}{2}\rangle$ is $\omega_{21} \approx 5$ kHz. By virtue of the selection rule $\Delta m_z = 1$, the transition between the sublevels occurs under the action of one of the circular components of the RF field. The frequency $\omega_{21}$ exceeds by many orders of magnitude the frequencies of the transitions to the nearest levels, so that such a system can be regarded with high degree of accuracy as a two-level system suitable for use as a model. However, because of the small population difference ($\approx 10^{-10}$) the observation of the absorption of the RF-field photons at the attainable $^{113}$Cd vapor concentrations ($10^{10} - 10^{11}$ cm$^{-3}$) is practically impossible. The population difference needed for the registration ($\approx 10^{-2}$) was therefore produced by the method of optical orientation.[11] Predominant population of one of the Zeeman levels of $^{113}$Cd produced resonant radiation ($\lambda = 3261$ Å) directed along $H$, from a lamp with the isotope $^{114}$Cd. The redistribution of the excitation over the Zeeman levels under the influence of the light is accompanied by some bleaching of the vapor at the frequency of this radiation. Any perturbation that upsets this population redistribution leads to modulation of the intensity of the optical beam registered by the photoreceiver, and the signal from the latter is fed to an oscilloscope.

In our experiments the frequency $\omega_{21}$ remained constant and the frequency of the effective radiation field varied linearly with time (Fig. 1). Such a realization of nonadiabatic passage leads to results identical with those obtained above.

The choice of the rate of change of the frequency $b$ was determined by two mutually exclusive requirements. First, the passage must be fast enough to prevent $x$ radiative relaxation from occurring in the system

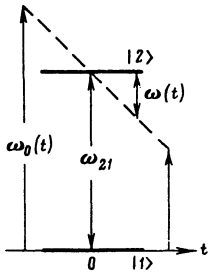


FIG. 1. Scheme of experimental realization of nonadiabatic passage in a two-level system.

during the time of passage. Under our conditions the lifetime of the excited state was $T = 0.09$ sec. Second, to be able to investigate the situation $\delta \gg 1$ while satisfying the condition (10), the passage must be slow enough, $b\tau_c^2 \ll 1$. The time $\tau_c$ defined by relation (9) could be varied by varying the width of the RF emission spectrum. The spectral composition of this radiation was formed with the aid of narrow-band rectangular filters through which a practically "white" shot noise passed. The width of the pass band of the filters $\Delta_w$ was much less than the width of the shot-noise. The value $\Delta_w = 10^3$ Hz was a compromise that limited the chosen rate of change of frequency to $b/2\pi = 1.7 \times 10^4$ Hz/sec.

In the experiment we registered the signal of the radiation field passing through a cell with $^{113}$Cd vapor and proportional to the change of the population difference after a single passage through the nonadiabaticity region. A direct comparison of this signal with the theoretically obtained quantities was impossible because of the appreciable fluctuations of the change of the population in the nonmonochromatic field (at a rather exact determination of the initial population difference, and at signal/noise = 50). We have therefore averaged the oscillograms of the signals by exposing them on photographic films. The number of exposures at a fixed radiation power was about 200. This number of realization made it possible to determine with sufficient accuracy not only the average value of the population change, but also their fluctuations, whose value amounted to ≈30%.

For a comparison with the situation of the excitation of the transition by broadband radiation, we investigated experimentally the excitation of the same system by a field with a moderate width of the spectrum, $\Delta_w = 100$ Hz ($b\tau_c^2 = 10.7$) and with monochromatic radiation. The obtained dependences of the transition probabilities on the field intensity are shown in Fig. 2. The same figure shows the results of the corresponding calculations performed without fit parameters in accordance with formulas (8) for $\Delta_w = 10^3$ Hz and (11) (see below) for a monochromatic field. It is seen that the results of the theory and of the experiments are in good agreement.

4. We now discuss the results. Despite the similarity between the obtained expression (8) and the Zener formula

$$w_м = 1 - e^{-\delta}, \tag{11}$$

which describe the transition probability following a single passage through the resonance point, the results of the action of intense ($\delta > 1$) monochromatic and nonmonochromatic fields are substantially different in the case of a double passage. Nonmonochromatic excitation equalizes the populations, since the probability of the transition in double passage is

$$W_w = {}^1/_2(1 - e^{-4\delta}). \tag{12}$$

In a monochromatic field, on the other hand, a double passage results in a decrease of the excitation

$$W_м = 2e^{-\delta}(1 - e^{-\delta}). \tag{13}$$

Another interesting feature of the result is that the change of the linear dependence of the excitation on the intensity into a nonlinear dependence, in both monochromatic and nonmochromatic fields, is characterized by the same parameter $\delta \approx 1$.

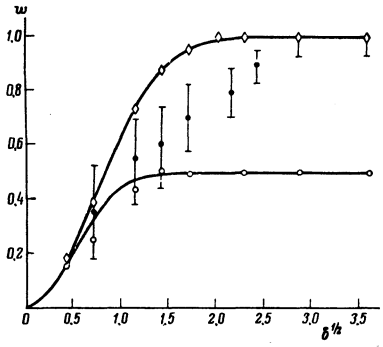


FIG. 2. Dependence of the transition probability $w$ on $\delta^{1/2} \sim (\langle E^2 \rangle)^{1/2}$ in single passage through resonance: ○—broad spectrum ($\Delta_w = 10^3$ Hz), ◇—monochromatic field, ●—spectrum width $\Delta_w = 10^2$ Hz. Solid lines—result of calculation by formulas (8) and (11). The dimension of the vertical lines determines the variances of the population.

Thus, to observe nonlinear optical phenomena when colliding atoms are nonresonantly excited there is no need to narrow down the spectrum of the effective radiation. The nonlinear phenomena due to the nonadiabatic passage through "resonance" are caused by the intensity of the total radiation spectrum, although, of course, the character of the dependence of these phenomena on the intensity is different for broad and narrow spectra of the applied radiation. In particular, to register the phenomenon of "bleaching of the medium without saturation" it is necessary that during the time of passage through the region of the nonadiabaticity the field frequency have not enough time to change, i.e., at a time of flight $\approx 10^{-12}$ sec the width of the spectrum must not exceed ≈30 cm$^{-1}$. Excitation of the medium in the region of the far wing of the atomic line by radiation of a broader spectral composition should lead to saturation so long as the condition $a/\Delta_w < 1$ is not violated .

In conclusion, the authors thand V. V. Khromov for a discussion of the work.

[1)] See Refs. 8 and 9 concerning this topic.
[2)] The quasistatic case is considered in a paper by Zusman.[10]

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# Diffusion mechanism of ionization of highly excited atoms in an alternating electromagnetic field

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A new mechanism is proposed for description of the ionization of highly excited atomic states by a strong low-frequency electromagnetic field. It consists of diffusion of the electron along atomic states highly perturbed by the field. The diffusion time is calculated, and the range of fields in which this mechanism is dominant is estimated. The results are compared with existing experimental data.

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## 1. INTRODUCTION

The ionization of atoms and molecules in a strong electromagnetic field differs substantially from the ionization process in a constant field as the result of the presence of an additional parameter–the field frequency $\omega$. It is well known[1] that the square of the ratio $\gamma$ between the external field frequency and the frequency $\omega_t$ of tunneling of the electron through the potential barrier is the dominant factor:

$$\gamma=\frac{\omega}{\omega_t}=\frac{\omega}{\mathscr{E}}(2E_n^{(0)})^{1/2},$$

where $\mathscr{E}$ is the external field strength and $E_n^{(0)}$ is the binding energy of the electron in the atom (here and below we use atomic units $m=\hbar=e=1$). The analytic solution of the problem of ionization of a particle bound by short-range forces, which was obtained by Keldysh[1] on the assumption that $\mathscr{E}\gg\mathscr{E}_{at}$ (where $\mathscr{E}_{at}=5\times10^9$ V/cm is the atomic field strength) and $E_n^{(0)}\gg\omega$, showed that for $\gamma^2>10$ the ionization process has a multiphoton nature, while for $\gamma^2<1$ it has the nature of a tunnel effect. The solution of the same problem obtained by other authors[2,3] for a circularly polarized field without the limitations on field strength and frequency mentioned above is essentially similar to the results obtained by Keldysh.

At the present time there is no general description of the process of ionization of a real atom in an alternating field. Data have been obtained only by means of perturbation theory for the multiphoton limiting case and by the quasiclassical method for the tunneling limiting case (see for example Ref. 4). Inclusion of the long-range nature of the Coulomb potential, carried out in terms of the quasiclassical method, did not result in a change in the value of $\gamma^2$ which marks the boundary between the tunnel and multiphoton regimes.[5]

However, separation of the ionization process into two limiting cases—tunneling and multiphoton—is valid only in a situation where the perturbation of the discrete atomic spectrum does not affect the transition probability. We note that, depending on the field strength and frequency of the ionizing field and also on the binding energy of the initial state, perturbation of the spectrum can play an important role for various values of the parameter $\gamma$ (excluding the tunneling case).

In what follows we shall discuss the hydrogen atom, since the excited states of interest to us are always hydrogenlike. The perturbation of a bound electronic state with principal quantum number $n$ is strong when the splitting of this state reaches a magnitude comparable with the distance to the closest shell, i.e., when we have the condition

$$\delta E_n/\Delta E_{n,\,n+1}\geqslant 1. \tag{1}$$

We note that the condition (1) is simultaneously a condition of inapplicability of the standard nonstationary perturbation-theory approach,[6] in which the unperturbed spectrum is taken as the basis.

It is an important circumstance that satisfaction of condition (1) is in no way equivalent to the condition for occurrence of the tunnel effect ($\gamma^2<1$) (Ref. 7). For this reason the problem of describing the ionization of an

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