Dynamic Stark effect in transitions from a discrete level to a continuum

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(Submitted 16 June 1981)
Zh. Eksp. Teor. Fiz. **81**, 1693–1700 (November 1981)

An analysis is made of the behavior of an isolated atom (or an impurity center in a semiconductor) in a strong electromagnetic field characterized by a photon energy slightly higher than the ionization potential of the atom (impurity center). It is shown that an increase in the effective ionizaton potential occurs in sufficiently strong fields so that one-photon absorption disappears. The role of two-photon transitions is discussed.

PACS numbers: 32.60. + i, 71.70.Ej

1. The Stark effect in an alternating field (dynamic Stark effect) has been investigated intensively in the case of two-level systems.[1] We shall consider the dynamic Stark effect in the case of transitions from a discrete level to a continuum. The transitions can occur in, for example, an atom or an impurity center in a semiconductor in a strong electromagnetic field of photon energy $\hbar\omega$ slightly higher than the ionization potential I of the atom or impurity center (Fig. 1a). We shall show that when the field is sufficiently strong, the effective ionization potential increases so that it becomes greater than the photon energy. One-photon absorption then disappears. Manifestation of this effect depends on the profile of the linear absorption edge. If the absorption rises from zero,[1)] there is a certain threshold or critical field I', beginning from which the effective ionization potential $\mathscr{E}_c$ becomes greater than $\hbar\omega$. One-photon absorption vanishes when the field reaches the threshold value. In the range $\mathscr{E} > \mathscr{E}_c$ a strong bound state is formed: it represents a superposition of states at the discrete level and states in the continuous spectrum, and this bound state is localized near the atom in a region $[\hbar^2/m(I'-\hbar\omega)]^{1/2}$. Under these conditions the absorption is entirely due to many-photon transitions.

The threshold field intensity decreases on increase in the steepness of the absorption edge and in the hypothetical limiting case of a step-like edge of the linear absorption process a localized state is formed in an electromagnetic field no matter how low the intensity of this field. If there is a system of excited levels of increasing density on approach to the continuum, the effect becomes much more complex. We shall discuss the system shown in Fig. 1 and ignore excited states.

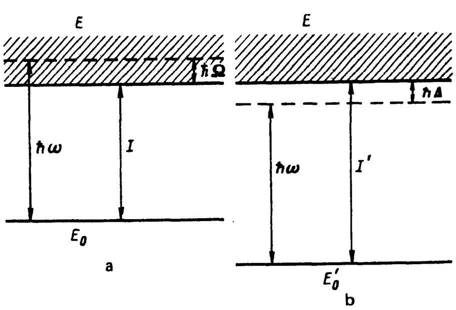

FIG. 1. a) Discrete level and continuum in a weak electromagnetic field with a photon energy $\hbar\omega$ not exceeding the ionization potential I. b) Discrete level and a continuum in a strong electromagnetic field of amplitude $\mathscr{E} > \mathscr{E}_c$.

2. The wave function of an electron in the presence of an electromagnetic wave should satisfy the Schrödinger equation

$$i\hbar\frac{\partial\psi}{\partial t} = (\hat H + \hat V)\psi. \tag{1}$$

Here, $\hat H$ is the Hamiltonian describing the stationary unperturbed states of an electron in atom; $\hat V$ is the operator of the interaction of an electron with the wave field, given by

$$\hat V = -\tfrac{1}{2}\hat d(\mathscr{E} e^{-i\omega t} + \mathscr{E}^* e^{i\omega t}), \tag{2}$$

and $\hat d$ is the projection of the dipole moment operator along the direction of the electric field vector $\mathscr{E}$ of the electromagnetic wave.

The wave function ψ will be sought in the form

$$\psi = B_0(t) e^{i\omega t}\psi_0 + \int_0^\infty B_E(t)\psi_E e^{-iEt/\hbar}\, dE. \tag{3}$$

Here ψ_0 is the wave function of an electron at a discrete level of energy $E_0 = -I$; ψ_E are the wave functions of electron states in a continuum, normalized to the δ function of the energy; E is the energy measured from the edge of the continuum. The integral in Eq. (3) implies, if necessary, also summation over other (apart from the energy) quantum numbers.

Equations (1)–(3) yield the following expressions for the coefficients $B_0(t)$ and $B_E(t)$:

$$\begin{gathered} i\hbar\frac{\partial B_0(t)}{\partial t} = \hbar\Omega B_0(t) - \frac{1}{2}(\mathscr{E} e^{-2i\omega t} + \mathscr{E}^*)\int_0^\infty d_{0E} B_E(t) e^{-iEt/\hbar}\, dE, \\ i\hbar\frac{\partial B_E(t)}{\partial t} = -\frac{1}{2}(\mathscr{E} + \mathscr{E}^* e^{2i\omega t}) d_{E0} B_0(t) e^{iEt/\hbar}. \end{gathered} \tag{4}$$

Here, $\hbar\Omega = \hbar\omega + E_0 = \hbar\omega - I$; d_{0E} is the matrix element of the dipole moment operator corresponding to a transition between a discrete level and a state in a continuous spectrum. In the system (4) we have ignored the terms describing transitions between the states in a continuous responsible for many-photon processes. The role of many-photon transitions will be discussed later.

We shall assume that $B_E = 0$ in the limit $t = -\infty$, express $B_E(t)$ in terms of $B_0(t)$ using the second equation

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in the system (4), and substitute the result into the first equation. We then obtain

$$B_E(t)=\frac{i}{2\hbar}\int_{-\infty}^{t}(\mathscr{E}+\mathscr{E}^*e^{2i\omega t'})d_{E0}B_0(t')e^{iEt'/\hbar}dt', \tag{5}$$

$$i\hbar\frac{\partial B_0(t)}{\partial t}=\hbar\Omega B_0(t)-\frac{|\mathscr{E}|^2}{4}\int_0^\infty B_0(t-\tau)K(\tau)d\tau,$$

$$K(\tau)=-\frac{i}{\hbar}(1+e^{-2i\omega\tau})\int_0^\infty|d_{E0}|^2e^{-iE\tau/\hbar}dE. \tag{6}$$

The terms oscillating at a frequency 2ω are dropped from the system (6).

3. The solution of the integral Eq. (6) can be found in the form $B_0(t)\propto e^{i\Delta t}$. The quantity Δ is described by

$$\Omega+\Delta=|\mathscr{E}|^2\tilde{\chi}(\Delta)/4\hbar; \tag{7}$$

$$\tilde{\chi}(\Delta)=\int_0^\infty K(\tau)e^{-i\Delta\tau}d\tau,$$

$$\tilde{\chi}(\Delta)=\int_0^\infty|d_{E0}|^2\left(\frac{1}{E+\hbar\Delta-i\nu}+\frac{1}{E+\hbar\Delta+2\hbar\omega-i\nu}\right)dE. \tag{8}$$

Here, $\nu\to0$.

Equation (7) defines the quasienergy E_0' of a discrete level perturbed by a field: $E_0'=E_0-\hbar\Delta-\hbar\Omega$ (Fig. 1b). The new "ionization potential" is

$$I'=I+\hbar\Delta+\hbar\Omega=\hbar\omega+\hbar\Delta.$$

We shall show that under certain conditions Eq. (7) has a real solution $\Delta>0$. Then, $I'>\hbar\omega$ and one-photon ionization does not occur. The function $\tilde{\chi}(\Delta)$ is identical with the linear polarizability at a frequency ω for an atom whose discrete level is shifted by an energy $\hbar\Delta+\hbar\Omega$ downward (this corresponds to the replacement of E_0 with E_0'). Apart from small terms $\sim\Omega/\omega$, we can say that $\tilde{\chi}(\Delta)$ is identical with the linear polarizability of an unperturbed atom $\chi(\omega')$ at a frequency ω', which is less than the field frequency ω by an amount $\Omega+\Delta$:

$$\tilde{\chi}(\Delta)=\chi(\omega-\Omega-\Delta)=\chi((I-\hbar\Delta)/\hbar). \tag{9}$$

We can see from the system (8) that the right-hand side of Eq. (7) decreases monotonically on increase in $\Delta>0$, whereas the left-hand side rises monotonically. Therefore, there is a solution $\Delta>0$ of Eq. (7) provided the right-hand side of Eq. (7) corresponding to $\Delta=0$ is greater than Ω.

If $|d_{E0}|^2$ is finite for $E=0$, it then follows that $\tilde{\chi}(\Delta)\to\infty$ in the limit $\Delta>0$ and the solution $\Delta>0$ exists for any value of the field intensity. Then, for low values of Δ we have

$$\tilde{\chi}(\Delta)=(|d_{E0}|^2)_{E=0}\ln(\omega_0/\Delta), \tag{10}$$

where ω_0 is the frequency of the atomic order defined by

$$\ln\frac{\omega_0}{\omega}=\frac{1}{(|d_{E0}|^2)_{E=0}}\int_0^\infty(|d_{E0}|^2)'\ln\frac{2\hbar^2\omega^2}{E(E+\hbar\omega)}dE.$$

The prime denotes differentiation with respect to E and within the approximations adopted here we can assume that $\hbar\omega=I$. Solution of Eq. (7) in low fields then has the form

$$\Delta=\omega_0\exp[-4\hbar\Omega/(|d_{E0}|^2)_{E=0}\mathscr{E}^2], \tag{11}$$

which resembles the expression for the Cooper coupling energy in the theory of superconductivity.[2)]

We shall now consider the case when $|d_{E0}|\to0$ in the limit $E\to0$. In this case the value of $\tilde{\chi}(\Delta)$ is finite and the solution of Eq. (7) for real values of $\Delta>0$ exists in fields exceeding the threshold or critical value $\mathscr{E}_c$ given by

$$\mathscr{E}_c^2=4\hbar\Omega/\tilde{\chi}(0). \tag{12}$$

If $\hbar\Omega\ll I$, which is true in the case under discussion, the threshold field is much less than the atomic value.

4. It is interesting to consider the behavior of Δ in fields lower than the threshold value. Then, Δ is a complex quantity given by $\Delta=\Delta'+i\Gamma$, where Γ is the decay of the bound state due to the ionization of an atom. We shall see below that $\Gamma\ll|\Delta'|$ and $\Delta'<0$ so that Eq. (8) yields

$$\tilde{\chi}(\Delta)=\tilde{\chi}(0)+i\pi(|d_{E0}|^2)_{E=-\hbar\Delta'}. \tag{13}$$

Then, Eq. (7) gives

$$\Omega+\Delta'=\tilde{\chi}(0)\frac{\mathscr{E}^2}{4\hbar},\quad \Gamma=\pi(|d_{E0}|^2)_{E=-\hbar\Delta'}\frac{\mathscr{E}^2}{4\hbar}. \tag{14}$$

It is clear from Eq. (14) that the shift and decay of the level are governed by the shape of the absorption edge, i.e., by the dependence of $|d_{E0}|^2$ on E at the threshold. We shall assume that at the absorption threshold we have $|d_{E0}|^2=\chi_0(E/I)^\alpha$, where χ_0 is a quantity of the order of $\tilde{\chi}(0)$ and α is a positive number. It then follows from the expressions in Eq. (14) that

$$\Delta'=\tilde{\chi}(0)\frac{\mathscr{E}^2}{4\hbar}-\Omega,\quad \Gamma=\pi\chi_0\frac{\mathscr{E}^2}{4\hbar I^\alpha}\left[\Omega-\tilde{\chi}(0)\frac{\mathscr{E}^2}{4\hbar}\right]^\alpha. \tag{15}$$

We can now see that when the field increases from zero the value of Δ' rises from $\Delta'=-\Omega$ (this corresponds to a shift of the level energy) and vanishes at the threshold field. The low-field decay Γ is proportional to $\mathscr{E}^2$ and reaches it maximum at $\mathscr{E}^2=\mathscr{E}_c^2/(1+\alpha)$ and then vanishes at the threshold field. The maximum value of Γ is of the order of $\Omega(\hbar\Omega/I)^\alpha$. Figure 2 shows schematically the field dependences of Δ' and Γ. By way of example, we shall consider the exactly soluble problem of a center with zero radius. We then have[3,4]

$$\alpha=3/2,\quad \chi_0=(16/3\pi)a_0^3,\quad \tilde{\chi}(0)=[(2^{11/2}-44)/3]a_0^3.$$

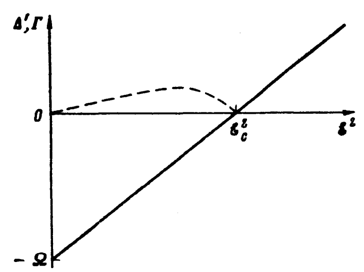

FIG. 2. Dependence of Δ' (continuous line) and of the decay constant (dashed curve) on the applied field.

Here, $a_0=(\hbar^2/2mI)^{1/2}$. In the case of a one-dimensional delta potential, we obtain

$$\alpha={}^1/_2,\quad \chi_0=(4/\pi)a_0^3,\quad \tilde{\chi}(0)=(7-2^{5/2})a_0^3.$$

5. We shall now consider the problem of the wave function of an electron in a quasibound state in a field exceeding the critical value. It follows from Eqs. (3) and (5) that if $B_0(t)\propto e^{i\Delta t}$, we obtain

$$\psi=Ae^{-iE_0't/\hbar}\left[\psi_0+\frac{\mathscr{E}}{2}e^{-i\omega t}\int_0^\infty\frac{\psi_E d_{E0}}{E+\hbar\Delta}dE+\frac{\mathscr{E}^*}{2}e^{i\omega t}\int_0^\infty\frac{\psi_E d_{E0}}{E+\hbar\Delta+2\hbar\omega}dE\right],\qquad(16)$$

where the quasienergy is $E_0'=-\hbar(\omega+\Delta)$. (We recall that zero on the energy scale is the boundary of the continuous spectrum.) We may conclude from Eq. (16) that the wave function is a superposition of a discrete state and of an electron cloud formed from the states in the continuous spectrum and having the dimensions $(\hbar^2/m\hbar\Delta)^{1/2}$ [this localization radius corresponds to the second term in the brackets in Eq. (16)]. The function (16) is normalizable and the normalization constant can be written in the form

$$A=(1+W)^{-1/2};\quad W=-\frac{\mathscr{E}^2}{4\hbar}\frac{d\tilde{\chi}(\Delta)}{d\Delta}.\qquad(17)$$

The quantity W gives the ratio of the probability of finding an electron in a cloud to the probability of finding it in a discrete state. If at the threshold we have $|d_{E0}|^2\propto E^\alpha$, then in the range $\alpha<1$ this ratio amounts to $\sim(\Omega/\Delta)^{1-\alpha}(\hbar\Omega/I)^\alpha$ (in a field higher than the threshold value). We can see that $W\to\infty$ in the limit $\Delta\to 0$, i.e., this occurs in the threshold field. However, even a slight excess above the threshold value causes W to decrease and become a small quantity ($\Delta=\Omega$ when $\mathscr{E}^2=2\mathscr{E}_c^2$). If $\alpha>1$, then $W\sim\hbar\Omega/I$ and it is always small.

We can easily calculate the average dipole moment in the state of Eq. (16) and find the effective permittivity of the medium governing the velocity of a strong wave:

$$B_0(t)|_{t\to\infty}=(1+W)^{-1},$$

where ε_0 is the contribution to the permittivity not associated with the transitions under discussion, N is the concentration of the atoms, and $\chi(\omega)$ is the atomic polarizability. If $\mathscr{E}>\mathscr{E}_c$ is real.

6. We shall consider the behavior of our system after abrupt application of an electromagnetic field at a moment $t=0$. We shall assume that $B_E=1$ for $t<0$. Then, we find that $B_0(t)$ is described by an integral equation differing from Eq. (6) only by the fact that the upper limit in the integral with respect to τ is t. This equation is easily solved by the Laplace transformation method and in the limit $t\to\infty$ only the contribution of the $S=i\Delta$ remains (we are assuming that the field is higher than the threshold value and Δ is real). We can easily show that in this case we have

$$\varepsilon=\varepsilon_0+4\pi N\chi(\omega-\Omega-\Delta),$$

where W is given by Eq. (17). Hence, it is clear that in a field which is not too high above the threshold an electron is most likely to remain in the state (16) and it will not be ejected from an atom.

When a field is applied abruptly, its spectrum has all the frequencies including those of the atomic order. A more interesting (and of greater practical importance) is the case when a field is applied slowly. We then find that $B_0(t)$ can be described by a differential equation. If we allow for the time dependence of the amplitude $\mathscr{E}$, we obtain the following equation instead of Eq. (6):

$$i\hbar\frac{\partial B_0(t)}{\partial t}=\hbar\Omega B_0(t)-\frac{\mathscr{E}(t)}{4}\int_0^\infty\mathscr{E}(t-\tau)B_0(t-\tau)K(\tau)d\tau.\qquad(18)$$

We shall make the substitution

$$B_0(t)=b_0(t)\exp\left(i\int_0^\infty\Delta(t')dt'\right),$$

where $\Delta(t)$ satisfies Eq. (7) for the value $\mathscr{E}$ at the moment in question; then, $b_0(t)$ is described by

$$i\hbar\frac{\partial b_0(t)}{\partial t}=-\frac{\mathscr{E}(t)}{4}\int_0^\infty K(\tau)e^{-i\Delta(t)\tau}[\mathscr{E}(t-\tau)b_0(t-\tau)e^{i\Phi}-\mathscr{E}(t)b_0(t)]d\tau,$$
$$\Phi=\Delta(t)\tau-\int_t^{t-\tau}\Delta(t')dt'\approx\frac{1}{2}\frac{d\Delta(t)}{dt}\tau^2.\qquad(19)$$

In the last case we have allowed for the fact that $\Delta(t)$ does not change greatly over time intervals on the atomic scale and the expression in the brackets of Eq. (19) can be expanded as a series in terms of τ and it is possible to retain only the terms containing the first derivatives with respect to time. Then, Eq. (19) becomes

$$\left[1-\frac{\mathscr{E}^2}{4\hbar}\frac{d\tilde{\chi}(\Delta)}{d\Delta}\right]\frac{\partial b_0(t)}{\partial t}=\left[\frac{\mathscr{E}^2}{8\hbar}\frac{d\Delta}{dt}\frac{d^2\tilde{\chi}(\Delta)}{d\Delta^2}+\frac{1}{8\hbar}\frac{d\mathscr{E}^2}{dt}\frac{d\tilde{\chi}(\Delta)}{d\Delta}\right]b_0(t).\qquad(20)$$

This equations is readily solved:

$$b_0=(1+W)^{-1/2}.$$

Here, W is described by Eq. (15) for instantaneous values of $\mathscr{E}^2$ and Δ. Since b_0 is identical with the normalization constant A in the wave function (16) the probability that an electron remains in a quasibound state up to t is

$$\frac{|B_0|^2}{|A|^2}=\exp\left[-2\int_0^t\Gamma(t')dt'\right],\quad\Gamma(t)=\mathrm{Im}\,\Delta(t).\qquad(21)$$

Equation (21) describes the decay of a quasibound state up to a moment t_c at which the field reaches its threshold value. The total decay probability in fields $\mathscr{E}(t)>\mathscr{E}_c$ is

$$1-\exp\left[-2\int_0^{t_c}\Gamma(t')dt'\right].\qquad(22)$$

The probability of decay is low if the atom is not ionized in a time that the field rises to the threshold value.

7. We shall now consider the role of two-photon absorption. The decay constant Γ_2 representing two-photon absorption is proportional to $\mathscr{E}^4$ and, therefore, its order of magnitude is $\mathscr{E}^4a_0^6/\hbar I$. If the field exceeds the critical value, we find that $\Gamma^2\sim\Omega(\hbar\Omega/I)$. On the other hand, the maximum value of the one-photon decay constant is $\Gamma\sim\Omega(\hbar\Omega/I)^\alpha$ [we recall that α is the power ex-

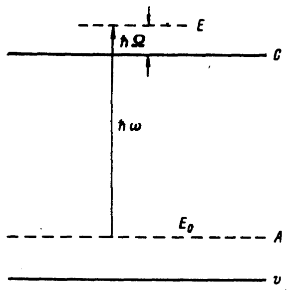

FIG. 3. Optical transition of an electron from an impurity level of an acceptor type (*A*) to the conduction band.

ponent in the dependence of the photoionization cross section on the excess of the photon energy above the ionization energy: $\sigma \sim (\hbar\omega - I)^{\alpha}$]. If $\alpha = \frac{3}{2}$, we find that, as in the case of negative hydrogen ions, two-photon absorption in fields of the order of the threshold value masks completely those changes in the one-photon absorption which are discussed above. However, if $\alpha = \frac{1}{2}$, then in a wide range of fields the two-photon absorption process should be much weaker than the one-photon process.

The value $\alpha = \frac{1}{2}$ corresponds to a hypothetical case of a one-dimensional potential and to a real case of an allowed transition between a deep center and a band state in a semiconductor. For example, it applies to a transition between an acceptor *p*-type state and an *s*-type conduction band, accompanied by the creation of a hole at an acceptor and of an electron in a band (Fig. 3).

We shall conclude by estimating the threshold or critical optical power. If $\hbar\Omega \sim 10$ meV and $a_0 \sim 30$ Å, we find that $\mathscr{E}_c \approx 4\hbar\Omega/a_0 \sim 4 \cdot 10^5$ V/cm, which corresponds to a power density of $\sim 10^9$ W/cm^2. The maximum value of the one-photon decay constant is $\sim 10^{12}$ sec^{-1} (for $\alpha = \frac{1}{2}$ and $I = 1$ eV) whereas the two-photon absorption is an order of magnitude less. Light pulses rising to the critical power density in a time shorter than Γ^{-1} should travel without absorption by the substance.

Photoionization of an atom in a strong field was considered by Kazakov *et al.*[5] using the general Fano theory[6] on the interaction of a discrete level with a continuous spectrum. Impurity-band electron transitions in the field of a strong electromagnetic wave were discussed by Elesin.[7] However, the treatments reported in Refs. 5 and 7 ignored not only the possibility that a level may split off from the continuous spectrum because of the dynamic Stark effect, but also the effective ionization potential.

1)An example of such a system is the negative hydrogen ion H^- or deep impurity centers in semiconductors.

2)The analogy between the problem of the interaction of a discrete level with a continuous spectrum (to which our problem is reduced) and the Cooper pairing was pointed out in the book by Baz', Zel'dovich, and Perelomov.[2]

3)In fact, since $\tilde{\chi}(\Delta)$ or its derivatives have a singularity at $\Delta \to 0$, the conditions of validity of Eq. (20) are more stringent: the field should not vary significantly in time intervals $\sim 1/\Delta$.

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Translated by A. Tybulewicz