

## THE EXCITATION AND IONIZATION OF ATOMS IN A STRONG RADIATION FIELD

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Submitted to JETP editor August 31, 1963

J. Exptl. Theoret. Phys. (U.S.S.R.) **46**, 1090-1097 (March, 1964)

Some general results are obtained regarding the behavior of atomic systems in a strong radiation field. By the latter is understood a radiation field with such a density that the energy of interaction between it and the atomic electrons approaches that between the electrons and nucleus. It is shown, in particular, that if the interaction time between the atom and field is sufficiently large, atomic ionization will be more probable than its excitation in the bound state, even though the field quantum  $\hbar\omega$  may be much smaller than the ionization potential  $I$ . Some estimates of the photoionization potential for  $\hbar\omega \ll I$  are presented.

1. It is clear from general considerations that the behavior of atomic systems in a strong radiation field, whose energy of interaction with the atomic electrons approaches the energy of interaction of the electrons in the nucleus, will differ qualitatively from the usual behavior predicted by the perturbation theory of quantum transitions. For these field intensities it is clear that the condition for resonance transitions derived in the first approximation of perturbation theory is violated. In the present case, from the point of view of perturbation theory, many photon processes become dominant and take place with the same probability as single-photon processes. If the interaction time of the atom with the radiation field is sufficiently long, then, as will be shown, the most probable process is the ionization of the atom (and not its excitation to a bound electron state), even though the quantum of the field  $\hbar\omega$  may be many times smaller than the ionization potential. An example of such a process is the much studied (both theoretically and experimentally) ionization of hydrogen atoms in a constant electric field (cf. for example [1,2]).

The critical values of the field strength, for which the above features of the quantum transitions begin to occur, may be considerably less than the values that correspond to the internal energy of the atom. The above example of the ionization of the hydrogen atom in a constant field supports this conclusion. As shown both by theory and experiment, the critical value of the field strength for ionization of the initial states of the components of the  $H_\gamma$  lines (principal quantum number  $n = 5$ ) is of order  $10^6$  V/cm, although the

interatomic field (for  $n = 5$ ) is of order <sup>1)</sup>  $10^8$  V/cm.

The radiation intensities obtainable from present day lasers approximate the above mentioned critical values <sup>2)</sup>. For a laser power  $P \sim 5 \times 10^2$  MW focused to a spot of diameter  $d \sim 10 \mu$ , the field strength  $\mathcal{E}_0 \approx 3 \times 10^8$  volts/cm. A rough estimate shows that for this laser power one should expect to ionize hydrogen atoms even in the ground state (for  $n = 1$  the strength of the interatomic field is of order  $5 \times 10^9$  volts/cm). The probability per unit time of ionizing this atom in a constant electric field is given by (in atomic units) [2]

$$w = (4/\mathcal{E}) \exp(-2/3\mathcal{E}). \quad (1)$$

Clearly we may use the same formula for calculating the ionization probability of a hydrogen atom in an alternating field of frequency  $\nu$ , replacing the quantity  $\mathcal{E}$  by the amplitude of this alternating field  $\mathcal{E}_0$ , if the probability  $w(\mathcal{E}_0)$  obtained in this way satisfies the condition  $w(\mathcal{E}_0) \gtrsim \nu$  (the adiabatic approximation). If this condition is satisfied the atom has the time to become ionized before the field  $\mathcal{E}_0 \cos 2\pi\nu t$  decreases significantly from its peak value. For a ruby laser  $\nu \approx 10^{-2}$  a.u.; for  $\mathcal{E}_0 \approx 3 \times 10^8 / 5 \times 10^9 = 6 \times 10^{-2}$

<sup>1)</sup>The value  $10^6$  V/cm corresponds to an ionization probability for a hydrogen atom per unit time of order  $10^8 \text{ sec}^{-1}$ , i.e., it is of order  $1/\tau_0$ , where  $\tau_0$  is the spontaneous lifetime of the level.

<sup>2)</sup>Since the present article was submitted for publication we have become aware of experimental observation<sup>[3]</sup> of the ionization of helium, argon and oxygen by ruby laser radiation.

a.u. Eq. (1) leads to a comparable value for the quantity  $w$  ( $\sim 10^{-2}$  atomic units). Thus  $w(\mathcal{E}_0)/\nu \sim 1$ , and one should expect ionization of the atom during a time of the order of the period of the radiation field ( $1/\nu$ ). This estimate of the critical value of the power  $P$  is clearly excessive because of the supplementary condition  $w/\nu \gtrsim 1$ , which clearly is not actually necessary.

2. At the radiation field intensities we are considering, the usual perturbation theory of quantum transitions becomes completely unsuitable. Hence in making a theoretical analysis of the behavior of atomic systems interacting with a strong field it is necessary to seek solutions to the equations of quantum mechanics which exhibit the total temporal behavior of the systems. In what follows we will consider the problem of quantum transitions in hydrogen-like atoms which are located in a monochromatic radiation field of circular polarization. This particular type of radiation is chosen for two reasons; first, it is possible in this case to obtain rigorously a whole set of interesting results; second, it is clear from physical considerations that several of these results are general in character and are valid for the case of a strong radiation field of arbitrary polarization (and, within wide limits, of arbitrary spectral composition).

Our treatment of the problem of the excitation of an atom in a circularly polarized radiation field involves transforming the problem to that of the excitation of the same atom in constant electric and magnetic fields. This transformation is very useful, since it permits formulation of the problem in terms of the familiar and well understood relations of the general theory of the spontaneous decay of quantum systems<sup>[4,5]</sup>.

3. We first consider the problem of quantum transitions in an atomic system (we are not requiring now that the system be hydrogen-like) under the influence of external, arbitrary, static fields. The total Hamiltonian of such a system is

$$H = H_0 + V, \quad (2)$$

where  $H_0$  is the Hamiltonian of the isolated atom and  $V$  is the time-independent Hamiltonian of the interaction with the external fields. We assume that at some initial instant  $t = 0$  this system is in some eigenstate  $\psi_{k_1}^{(0)}(q)$  of the Hamiltonian  $H_0$ ,

corresponding to a discrete energy level  $E_{k_1}^{(0)}$

( $q$  is the set of all coordinates of the atom). We wish to calculate the probability  $P_{k_1 k_2}(t)$  of a transition of the atom in time  $t$ , due to the effect of  $V$ , to the state  $\psi_{k_2}^{(0)}(q)$ , described by the set

of quantum numbers  $k_2$ . The final state  $k_2$  may belong either to a discrete energy spectrum (an excitation of the atom to a bound state) or to the continuous spectrum (ionization of the atom).

It follows from the general theory of spontaneous decay<sup>[4,5]</sup> that the initial state of the system we are considering will break up eventually (i.e., the atom will be ionized) only when the perturbation  $V$  is such as to change the energy spectrum of the system from discrete to continuous. More exactly, in order for such a process to occur, it is necessary and sufficient that the total Hamiltonian  $H$  have a continuous energy spectrum. In this case the functions giving the energy density distributions  $f_{k_1}(E) = |c_{k_1}(E)|$  and  $f_{k_2}(E) = |c_{k_2}(E)|^2$  in the initial and final states of the system do not contain singularities of the type<sup>3)</sup>  $\delta(E - E_0)$ ; we have

$$c_{k_{1,2}}(E) = \int \psi_E^*(q) \psi_{k_{1,2}}^{(0)}(q) dq, \quad (3)$$

where  $\psi_E(q)$  is an eigenfunction of the operator<sup>4)</sup>  $H$ .

It should be pointed out that the perturbation  $V$  due to a uniform electric field ( $V = -ex\mathcal{E}$ ) always satisfies the above requirement, i.e., it always spreads out the discrete energy spectrum of the atom into a continuous spectrum<sup>[2]</sup>. This fact in turn is reflected by the phenomenon of the ionization of atoms in such a field.

In general the transition probability  $P_{k_1 k_2}(t)$  is given by

$$P_{k_1 k_2}(t) = |a_{k_1 k_2}(t)|^2, \quad P_{k_1 k_2}(0) = 0, \quad (4)$$

where  $a_{k_1 k_2}(t)$  is the projection of the state

$$\Psi_{k_1}(q, t) = \exp(-iHt/\hbar) \psi_{k_1}^{(0)}(q)$$

of the system at the instant  $t$  on the state  $\psi_{k_2}^{(0)}(q)$ :

$$a_{k_1 k_2}(t) = \int \Psi_{k_1}(q, t) \psi_{k_2}^{(0)*}(q) dq = \int c_{k_1}(E) c_{k_2}^*(E) e^{-iEt/\hbar} dE. \quad (5)$$

Equations (4) and (5) give the following important result. The transition probability of the atom

<sup>3)</sup>In mathematical terms the above necessary and sufficient condition follows from the continuity of the integral of the distribution function

$$F_k(E) = F_k(E') + \int_{E'}^E f_k(E_1) dE_1.$$

<sup>4)</sup>The fact that the states of the continuous spectrum are practically always degenerate is not important to us, here, and hence we will not explicitly write the other quantum numbers which, along with the energy  $E$ , form a complete set.

$P_{k_1 k_2}(t)$  to a bound state (i.e., to a state corresponding to a discrete energy value  $E_{k_2}$  of the Hamiltonian  $H_0$ ) has at least one maximum and tends to zero as  $t \rightarrow \infty$ . This conclusion is not valid for a transition of the atom to an ionized state; for such a transition the probability  $P_{k_1 k_2}(t)$  in general does not approach zero as  $t \rightarrow \infty$  (the total probability for ionization always approaches unity). Physically this difference in the asymptotic behavior of the probabilities  $P_{k_1 k_2}(t)$  means that for a sufficiently large interaction time  $t$  between the atom and the perturbing field, the only probable process is the ionization of the atom.

This result may be easily understood on physical grounds which are also relevant to the case of the strong radiation field. The effect on an atom of a field which is strong enough to cause ionization is to cause a significant modification (distortion) of the electron cloud. If the interaction time with the field is not too long, the electron cloud of the atom is not totally destroyed and the probability of the atom making a transition to a discrete state of the Hamiltonian  $H_0$  is non-zero. As the interaction time is increased, the electron cloud is perturbed so strongly that its configuration begins to correspond only to an ionized state of the Hamiltonian  $H_0$  (this state continues for infinitely long  $t$ ).

The validity of this assertion follows from (4) and (5) and the theorem of Riemann-Lebesgue<sup>[6]</sup>, according to which the integral (5) approaches zero as  $t \rightarrow \infty$  if the real and imaginary parts of the function  $c_{k_1}(E) c_{k_2}^*(E)$  are absolutely integrable over the whole range of  $E$ . It is easy to show that this latter condition is always satisfied for discrete states  $\psi_{k_1}^{(0)}(q)$  and  $\psi_{k_2}^{(0)}(q)$ . In fact for such states one has

$$\int |c_{k_{1,2}}(E)|^2 dE < \infty, \quad (6)$$

i.e., the integral always converges (and is usually normalized to unity). The question of the absolute integrability of the functions

$$\varphi_1(E) \equiv \operatorname{Re} c_{k_1}(E) c_{k_2}^*(E) \quad \text{and} \quad \varphi_2(E) \equiv \operatorname{Im} c_{k_1}(E) c_{k_2}^*(E),$$

is clearly determined by the behavior of these functions for large values of  $E$ . However from the condition for the integrability of the functions  $(\operatorname{Re} c_{k_{1,2}}(E))^2$  and  $(\operatorname{Im} c_{k_{1,2}}(E))^2$  [which follows from (6)], it is clear that as  $|E| \rightarrow \infty$  the functions  $|\varphi_1(E)|$  and  $|\varphi_2(E)|$  approach zero at least as rapidly as  $|E|^{-\alpha}$ , where  $\alpha > 1$ , i.e., these functions are in fact absolutely integrable.

If, however, even one of the states  $k_1$  and  $k_2$  corresponds to the continuous spectrum of the Hamiltonian  $H_0$ , condition (6) is not fulfilled (the integral diverges), and the probability  $p_{k_1 k_2}(t)$ , in general, does not tend to zero. The above discussion shows that since the total transition probability to discrete states of the atom goes to zero, the total ionization probability  $W_{k_1}(t)$ , i.e., the total probability of ionization to all states of the continuous spectrum, must approach unity as  $t \rightarrow \infty$ . Thus for sufficiently long interaction times  $t$  the probability  $W_{k_1}(t)$  must coincide with the probability for the spontaneous decay of the initial state of the system, i.e.,<sup>5)</sup>

$$W_{k_1}(t) = 1 - \left| \int f_{k_1}(E) e^{-iEt/\hbar} dE \right|^2. \quad (7)$$

The ionization probability per unit time  $w_{k_1}$  is determined by the width of the energy level  $E_{k_1}^{(0)}$ .

More precisely, if the energy density distribution is given by the dispersion formula

$$f_{k_1}(E) = A / [(E - E_{k_1}^{(0)})^2 + \Gamma_{k_1}^2], \quad (8)$$

then<sup>6)</sup>

$$w_{k_1} = 2\Gamma_{k_1}/\hbar.$$

4. We now turn to the problem of quantum transitions in hydrogen-like atoms (and in particular in hydrogen atoms), interacting with a strong monochromatic radiation field of circular polarization in the  $(x, y)$  plane. The interaction between the optical electron of the atom and the field will be assumed to have a dipole character. Our treatment will be semiclassical in nature (the field will not be quantized); the transition probabilities calculated below will apply only to stimulated transitions in a strong field.

The Schrödinger equation describing the time dependence of the state  $\Psi(\mathbf{r}, t)$  of the optical electron has the form

$$i\hbar \partial\Psi/\partial t = \{H_0 - e\mathcal{E}_0(x \cos \omega t - y \sin \omega t)\} \Psi, \\ H_0 = -\frac{\hbar^2}{2\mu} \nabla^2 + U(r), \quad (9)$$

where  $H_0$  is the Hamiltonian of the isolated atom, and  $\mathcal{E}_0$  is the amplitude of the field. At  $t = 0$  the

<sup>5)</sup>Expression (7) is well known as the Fock-Krylov theorem.

<sup>6)</sup>The concept of time-independent transition probabilities (in particular, ionization probabilities) per unit time can, as is well known, be introduced only for sufficiently large  $t$  (strictly speaking for  $t \rightarrow \infty$ ). When this is true, (7) is always valid and also (in the most general case) we can use the dispersion formula (8)<sup>[4]</sup>.

atom is in the state  $\Psi(\mathbf{r}, 0) = \psi_{n_1 l_1 m_1}^{(0)}(\mathbf{r})$ , characterized by the values of the principal quantum number  $n_1$ , the azimuthal quantum number  $l_1$ , and the magnetic quantum number  $m_1$ .

We now transform to a new wave function

$$\Psi_r(\mathbf{r}, t) = \exp(i\omega t L_z) \Psi(\mathbf{r}, t), \quad (10)$$

where  $L_z$  is the operator of the  $z$ -component of the momentum of the electron; the operator  $\exp(i\omega t L_z)$  is the operator for rotation around the  $z$  axis by an angle  $\omega t$ . Clearly,

$$\Psi_r(\mathbf{r}, 0) = \Psi(\mathbf{r}, 0) = \psi_{n_1 l_1 m_1}^{(0)}(\mathbf{r}). \quad (11)$$

The equation for  $\Psi(\mathbf{r}, t)$  has the form<sup>7)</sup>

$$i\hbar \partial \Psi_r / \partial t = H_r \Psi_r, \quad H_r \equiv H_0 - \hbar\omega L_z - e\mathcal{E}_0 x. \quad (12)$$

Thus in a rotating coordinate system the problem of quantum transitions is equivalent to the problem treated above of quantum transitions under the influence of the static perturbation

$$V_r = -\hbar\omega L_z - e\mathcal{E}_0 x. \quad (13)$$

Clearly, this type of perturbation corresponds to the presence of mutually perpendicular constant electric and magnetic fields  $\mathcal{E}_0$  and  $\mathcal{H}_0 = \omega / (|e|/2\mu c)$  (along the  $x$  and  $z$  axes respectively).<sup>8)</sup> It is easy to show that the transformation to the rotating coordinate system does not alter the conclusions of the previous section. In particular the probability  $P_{n_1 l_1 m_1}^{n_2 l_2 m_2}(t)$  for an atomic transition from the state  $k_1 = (n_1, l_1, m_1)$  to the state  $k_2 = (n_2, l_2, m_2)$  during the time  $t$  is equal to

$$P_{n_1 l_1 m_1}^{n_2 l_2 m_2}(t) = |a_{n_1 l_1 m_1}^{n_2 l_2 m_2}(t)|^2, \quad (14)$$

where

$$a_{n_1 l_1 m_1}^{n_2 l_2 m_2}(t) = e^{-im_2 \omega t} \int c_{n_1 l_1 m_1}(E) c_{n_2 l_2 m_2}^*(E) e^{-iEt/\hbar} dE, \quad (15)$$

$$c_{nlm}(E) = \int \psi_E^{(r)*}(\mathbf{r}) \psi_{nlm}^{(0)}(\mathbf{r}) d\mathbf{r}, \quad (16)$$

where  $\psi_E^{(r)}(\mathbf{r})$  is an eigenfunction of the operator  $H_r$ . The expression (15) differs in form from expression (5) only by the factor  $\exp(-im_2 \omega t)$ , which does not effect the probability  $P_{n_1 l_1 m_1}^{n_2 l_2 m_2}(t)$ . It must be emphasized that this result was obtained only for a hydrogen-like atom ( $U(r)$  is a potential with central symmetry).

Since the perturbation  $V_r$  contains the electric field  $\mathcal{E}_0$ , the Hamiltonian  $H_r$  has a continuous energy spectrum (see the preceding section) which

in turn ensures the above-mentioned asymptotic properties for the transition probability  $P_{n_1 l_1 m_1}^{n_2 l_2 m_2}(t)$ .

In particular the total atomic ionization probability  $W_{n_1 l_1 m_1}(t)$  for a sufficiently long time  $t$  must be given by equation (7) for an arbitrary value of the frequency  $\omega$ . This probability always tends to unity with increasing  $t$ , i.e., the photoelectric effect can occur for a radiation field of any frequency if it is sufficiently strong.

It is interesting to compare these results with the known first-order perturbation theory results and the theory of spectroscopic saturation. It is easily seen that to a first approximation in the field equation (15) leads to the well known expression for the coefficient  $a_{kn}^{(1)}(t)$  in the expansion of the wave functions under a monochromatic perturbation<sup>[2] 9)</sup>. As far as the saturation effect is concerned, it follows from our considerations that the well known<sup>[2]</sup> description of this effect for atomic transitions from the state  $k_1$  to the state  $k_2$  and back at frequency  $\mu_{k_1 k_2} \mathcal{E}_0 / \hbar$  ( $\mu_{k_1 k_2}$  is the matrix element of the transition) remains valid only for values of the field which are not too large; that is, it remains valid as long as

$$w_{k_1, 2}(\mathcal{E}_0) \ll \mu_{k_1 k_2} \mathcal{E}_0 / \hbar, \quad (17)$$

where  $w_{k_1, 2}(\mathcal{E}_0)$  is the probability per unit time for ionization of the atoms in the states  $k_1$  and  $k_2$ . When this condition does not hold, the concept of saturation clearly loses its meaning.

5. The fact that the effect of a strong radiation field of arbitrary frequency on an atom is to cause the same physical result, i.e., ionization, as the effect of a strong constant electric field, makes it possible to introduce the concept of an effective ionizing field. By the latter we mean that constant electric field  $\mathcal{E}_{\text{eff}}$  which, when acting on a given atom in a given quantum state  $k_1$ , causes its ionization with the same probability  $w_{k_1}$  (per unit time) as the effect of the given radiation field of amplitude  $\mathcal{E}_0$  and frequency  $\omega$ . Clearly  $\mathcal{E}_{\text{eff}}$  depends on  $\mathcal{E}_0$  and  $\omega$ , and the particular form of this dependence (for a given atom) is determined in general by the initial state  $k_1$  and by the polarization of the radiation. It may be shown that in principle the field  $\mathcal{E}_{\text{eff}}$  may be found by an independent method, i.e., without solving the problem of the ionization probability in the given radiation field.

Introduction of the concept of the effective ionizing field is very advantageous since the problem

<sup>7)</sup>Here we have used the obvious equality  $\exp(i\omega t L_z) (x \cos \omega t - y \sin \omega t) \exp(-i\omega t L_z) = x$ .

<sup>8)</sup>We are not taking account of the electron spin.

<sup>9)</sup>One must calculate the coefficients  $c_{nlm}(E)$  using the initial Hamiltonian  $H_0 - \hbar\omega L_z$  and the selection rule  $|m_1 - m_2| = 1$ .

of ionization in a constant electric field is considerably simpler than the problem in the radiation field, since the former can be formulated simply as a problem of the elementary tunneling effect. For a hydrogen atom this problem has, as is well known, been solved completely [1,2,7].

We will now show how to find  $\mathcal{E}_{\text{eff}}$  for the above case of a radiation field with circular polarization; however we will limit ourselves to finding the first approximation in  $\omega$  to the value  $\mathcal{E}_{\text{eff}} = \mathcal{E}_0$ , corresponding to the adiabatic approximation (see Sec. 1). It is convenient in making the calculation to introduce the projection operator  $Q_{k_1}$ , which causes projection onto the initial atomic state  $\psi_{k_1}^{(0)}(\mathbf{r})$  ( $k_1 = (n_1, l_1, m_1)$ ).

In applying Eq. (7) to the present problem we may write it in the form (cf. [5])

$$W_{k_1}(t) = 1 - \text{Sp } Q_{k_1} Q_{k_1}^{(r)}(t); \tag{18}$$

$$Q_{k_1}^{(r)}(t) = e^{iH_r t/\hbar} Q_{k_1} e^{-iH_r t/\hbar}.$$

According to (12) the operator  $H_r$  may be written as follows:

$$H_r = H^{(0)} - \hbar\omega L_z - ex(\mathcal{E}_0 - \mathcal{E}_{\text{eff}});$$

$$H^{(0)} \equiv H_0 - ex\mathcal{E}_{\text{eff}}. \tag{19}$$

For sufficiently low frequencies  $\omega$  the two last terms in the expression for  $H_r$  may be treated as perturbations. Putting (19) in (18) and using the expansion for the S-matrix accurate to terms of order  $\sim \omega$ , we find

$$W_{k_1}(t) = W_{k_1}^{(0)}(t) + \frac{1}{i\hbar} \int_0^t dt_1 \text{Sp } Q_{k_1} \{e(\mathcal{E}_0 - \mathcal{E}_{\text{eff}}) [x, Q_{k_1}^{(0)}(t_1) + \hbar\omega [L_z, Q_{k_1}^{(0)}(t_1)]\}, \tag{20}$$

$$Q_{k_1}^{(0)}(t) = e^{iH^{(0)}t/\hbar} Q_{k_1} e^{-iH^{(0)}t/\hbar},$$

$$W_{k_1}^{(0)}(t) = 1 - \text{Sp } Q_{k_1} Q_{k_1}^{(0)}(t). \tag{21}$$

Here  $W_{k_1}^{(0)}$  is the ionization probability in the field  $\mathcal{E}_{\text{eff}}$ . The value of the quantity  $\mathcal{E}_{\text{eff}}$  is determined from this expression by imposing the requirement

that the first order terms approach zero as  $t \rightarrow \infty$ . In this way we obtain

$$\mathcal{E}_{\text{eff}} = \mathcal{E}_0 - \frac{\hbar\omega}{|e|} \frac{\int_0^\infty dt \text{Sp } Q_{k_1} [L_z, Q_{k_1}^{(0)}(t)]}{\int_0^\infty dt \text{Sp } Q_{k_1} [x, Q_{k_1}^{(0)}(t)]}. \tag{22}$$

In calculating the operator  $Q_{k_1}^{(0)}(t)$  for use in this formula it is of course possible to set  $\mathcal{E}_{\text{eff}} = \mathcal{E}_0$  [cf. (21) and (19)]. For a hydrogen atom the eigenfunctions  $\psi_E(\mathbf{r})$  of the operator  $(H_0 - ex\mathcal{E}_{\text{eff}})$  are well known [1,7] and the integrals in (22) can be evaluated. In this case it should be kept in mind that the matrix elements of the operator  $Q_{k_1}$  in the representation formed by the functions  $\psi_E(\mathbf{r})$ , as may easily be shown, are equal to  $c_{k_1}(E)c_{k_1}^*(E')$ . In estimating the order of magnitude of the second term in (22) one may clearly consider the ratio of integrals occurring in the latter expression to be of order  $1/a$ , where  $a$  is the size of the atom.

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