Multiphoton transitions in a hydrogenlike atom

V. P. Oleĭnik and M. M. Chumachkova

Semiconductors Institute, Ukrainian Academy of Sciences

(Submitted April 8, 1974)

Zh. Eksp. Teor. Fiz. 67, 926–929 (September 1974)

A formula is derived and investigated for the probability of an electromagnetic-field-induced multiphoton transition of a hydrogenlike atom from the ground state to the first-excited state. The inducing field consists of an intense electromagnetic wave of frequency $\omega_1 \ll (\epsilon_1-\epsilon_0)\hbar$ and weak electromagnetic radiation of frequencies $\omega_2 \sim (\epsilon_1-\epsilon_0)/\hbar$ (ϵ_0 and ϵ_1 are the energies of the ground and first-excited state of the atom).

The behavior of a quantum system in a strong electromagnetic field is determined to a considerable degree by the multiphoton processes that occur in it[1, 2], and whose role increases with increasing field intensity. The multiphoton processes lead, in particular, to a shift of the atomic levels[3], to a radical restructuring of the continuous energy spectrum of the system[4], and to the appearance of a number of physical effects[4, 5] due to the exclusively nonlinear character of the interaction of the electromagnetic radiation with the medium. In this paper we consider a multiphoton transition of a hydrogen like atom from the ground state to the first excited state under the magnetic wave of frequency ω_1 (laser source) and weak electromagnetic radiation with frequencies $\omega \neq \omega_1$. The calculation was performed within the framework of the relativistic theory of the quantum transitions (see, e.g.,[6]).

The Hamiltonian of the interaction of the valence electron of the atom with the electromagnetic field is given by

$$H_{int}=e_0\int d\mathbf{r}\,\Psi^+(\mathbf{r})\boldsymbol{\alpha}\mathbf{A}(\mathbf{r})\Psi(\mathbf{r}),\qquad e_0=|e|,$$
$$\mathbf{A}(\mathbf{r})=\sum_{\mathbf{k}\lambda}(2\omega V)^{-1/2}\mathbf{e}_\lambda(b_{\mathbf{k}\lambda}e^{i\mathbf{kr}}+b_{\mathbf{k}\lambda}^+e^{-i\mathbf{rk}}),\qquad (1)$$

where $\Psi(\mathbf{r})$ and $\mathbf{A}(\mathbf{r})$ are respectively the operators of the electronic and electromagnetic fields in the Schrödinger representation; $b_{\mathbf{k}\lambda}$ ($b_{\mathbf{k}\lambda}^+$) is the annihilation (creation) operator of a photon with momentum $\mathbf{k}$ and polarization λ; $\mathbf{e}_\lambda$ is a unit vector of the polarization; $\boldsymbol{\alpha}$ is the Diract matrix, and V is the volume of the principal region. We shall henceforth be interested only in transitions of the atom from the ground to the first excited state, and therefore in the expansion of the operator $\Psi(\mathbf{r})$ in a complete system of Coulomb wave functions it suffices to take into account only the wave functions $\varphi_{im}(\mathbf{r})$ which describe the states $1s_{1/2}(\varphi_{0m})$, $2s_{1/2}(\varphi_{1m})$ $2p_{1/2}(\varphi_{2m})$ and $2p_{3/2}(\varphi_{3m'})$:

$$\Psi(\mathbf{r})=\sum_{im}a_{im}\varphi_{im}(\mathbf{r}),\qquad (2)$$

where a_{im} is the operator of electron annihilation in the state i and m; m is the projection of the total angular momentum on the quantization axis, $m = \pm 1/2$; $m' = \pm 1/2, \pm 3/2$. We assume also that the following inequalities are satisfied

$$\varepsilon_3-\varepsilon_2\ll\omega_1\ll\varepsilon_1-\varepsilon_0$$

and neglect the quantity $(\epsilon_3-\epsilon_1)/(\epsilon_2-\epsilon_0) \sim (Z\alpha)^2$, assuming that $\epsilon_1 = \epsilon_2 = \epsilon_3$ (ϵ_i is the electron energy in the state φ_{im}, Ze_0 is the electric charge of the nucleus of the atom, and α is the fine-structure constant).

Using the usual theory of quantum transitions and confining ourselves only to terms of first order in the interaction with the weak field, we obtain the following expression for the amplitude of the transition 0, m → i, m′ with absorption of a photon of frequency ω_2:

$$M_{im'}=-i(\omega_1/\omega_2)^{1/2}\int_{T_0}^{T}dt\,e^{-i\omega_2t}\langle 0',\omega_1|a_{im'}e^{i(t-T)H}A_2e^{-i(t-T_0)H}a_{0m}^+|0,\omega_1\rangle\qquad (3)$$

($T_0 \to -\infty$, $T \to +\infty$). In this formula, $|0, \omega_1\rangle$ and $|0', \omega_1\rangle$, are ket-vectors that describe the state of the field of the laser photons in the initial and final instants of time respectively and the ground state of the electrons and photons with frequencies $\omega \neq \omega_1$. The remaining notation is:

$$H=A_1+\omega_1b^+b+(b^++b)A_2,\qquad b=b_{\mathbf{k}z}|_{\omega=\omega_1},$$
$$A_1=\sum_{im}\varepsilon_ia_{im}^+a_{im},$$
$$A_2=\sum_m[\beta_m(a_{0m}^+a_{2m}-a_{2m}^+a_{0m})+\mu(a_{0m}^+a_{3m}-a_{3m}^+a_{0m})],$$
$$\beta_m=\frac{m}{|m|}\frac{e_0}{(2\omega_1V)^{1/2}}\tilde\beta,$$
$$\tilde\beta=\int d\mathbf{r}\varphi_{0,1/2}^+(\mathbf{r})\alpha_z\varphi_{2,1/2}(\mathbf{r})=-iZ\alpha\frac{16}{81}\left(\frac{2}{3}\right)^{1/2},$$
$$\mu=\frac{e_0}{(2\omega_1V)^{1/2}}\tilde\mu_0,\qquad \tilde\mu_k=\int d\mathbf{r}\varphi_{k,1/2}^+(\mathbf{r})\alpha_z\varphi_{3,1/2}(\mathbf{r}),$$
$$\tilde\mu_0=-iZ\alpha\frac{32}{81}3^{-1/2}\qquad (k=0,1).\qquad (4)$$

In the derivation of (3) we used relativistic Coulomb functions of the hydrogenlike atom and the dipole approximation, and took into account, for simplicity, only the z-polarization of the photons; in addition, we have neglected the quantity $\tilde\mu_1$ ($\tilde\mu_1 \sim \tilde\mu_0 (Z\alpha)^2$. Formula (3) takes into account exactly the interaction of the lowest energy levels of the atom with the intense electromagnetic field.

The photon-absorption probability summed over the final states and averaged over the initial states of the electron and the field of the laser photons is given by

$$W={}^1/_2\sum_{m=\pm 1/2}\frac{\omega_1}{\omega_2}\int_{-\infty}^{+\infty}dt_1\int_{-\infty}^{+\infty}dt_2\exp[i\omega_2(t_2-t_1)]$$
$$\times\operatorname{Sp}\rho_{\omega_1}\langle 0|a_{0m}A_2(t_2)A_2(t_1)a_{0m}^+|0\rangle,\qquad A_2(t)\equiv e^{itH}A_2e^{-itH},\qquad (5)$$

$|0\rangle$ is the ground state of the electrons, ρ_{ω_1} is a statistical operator describing the electromagnetic field of the laser source.

To calculate the mean value over the fermion operators in the expression (5), we carry out a unitary transformation with the aid of the operator

$$F=\exp[D_2(b+b^+)]\exp[D_1(b-b^+)],$$

Choosing the operators D_2 and D_1 to satisfy the condition that the terms linear in the operators b and b^+ vanish from the transformed Hamiltonian FHF^{-1}. This condition yields (in the limit as $V \to \infty$)

$$D_1=n_1A_2, \quad D_2=n_2A_3,$$

$$A_3=\sum_{m=\pm 1/2}[\beta_m(a_{0m}^+a_{2m}+a_{2m}^+a_{0m})+\mu(a_{0m}^+a_{3m}+a_{3m}^+a_{0m})].$$

$$n_1=-\frac{\omega_1}{\omega_1^2-(\varepsilon_1-\varepsilon_0)^2}, \quad n_2=\frac{\varepsilon_1-\varepsilon_0}{\omega_1^2-(\varepsilon_1-\varepsilon_0)^2}. \tag{6}$$

If the inequality

$$\zeta=4|n_2|\left(V\frac{\bar{E}^2}{\omega_1}|\beta_m^2+\mu^2|\right)^{1/2}\ll 1 \tag{7}$$

is satisfied (E is the time-averaged electric component of the intensity of the strong electromagnetic field) we can confine ourselves in the expression for FHF^{-1} to the terms quadratic in the operators b and b^+. As a result we obtain

$$FHF^{-1}\approx A_1+\omega_1b^+b+n_2^2(\varepsilon_1-\varepsilon_0)(b+b^+)^2A_4,$$

$$A_4=\sum_{m=\pm 1/2}[(\beta_m^2+\mu^2)a_{0m}^+a_{0m}-\beta_m^2a_{2m}^+a_{2m}-\mu^2a_{3m}^+a_{3m}+\mu\beta_m(a_{3m}a_{2m}^++a_{2m}a_{3m+})]. \tag{8}$$

In the derivation of this formula we have neglected the terms n_1^2 in comparison with $n_2^2(\omega_1^2/(\epsilon_1-\epsilon_0)^2 \ll 1)$. Our unitary transformation is equivalent to summation of an infinite sequence of terms of the perturbation-theory series[7].

We note the following commutation relations for the operators A_i, allowance for which greatly facilitates the subsequent operations:

$$[A_2,A_1]=(\varepsilon_1-\varepsilon_0)A_3, \quad [A_3,A_1]=(\varepsilon_1-\varepsilon_0)A_2, \quad [A_4,A_1]=0,$$
$$[A_4,A_2]=2(\beta_m^2+\mu^2)A_3, \quad [A_4,A_3]=2(\beta_m^2+\mu^2)A_2.$$

Using now the indicated unitary transformation and choosing the statistical operator ρ_{ω_1} in the form

$$\rho_{\omega_1}=\frac{1}{2\pi}\int d^2\xi\frac{1}{|\xi|}\delta\left(|\xi|-\left(V\frac{\bar{E}^2}{\omega_1}\right)^{1/2}\right)|\xi\rangle\langle\xi|,$$

($|\xi\rangle$ is the ket-vector of the coherent state: $b|\xi\rangle=\xi|\xi\rangle$), we easily obtain the following formula for the probability of absorption, per unit time, of light with frequency lying in the interval ω_2, $\omega_2+d\omega_2$ (in ordinary units);

$$dW=2\pi\left(\frac{16}{81}\right)^2(Z\alpha)^2\frac{(e_0c)^2}{\hbar\omega_2}N(\omega_2)d\omega_2 \times\sum_{s=0,\pm1\ldots}J_s^2(\varkappa)\delta\left(\frac{\varepsilon_1-\varepsilon_0}{\hbar}+2\omega_1(s+\varkappa)-\omega_2\right). \tag{9}$$

Here $N(\omega)d\omega$ is the number of photons in the unit volume with frequencies in the interval ω, $\omega+d\omega$,

$$\varkappa=\frac{1}{8}\zeta^2\frac{\varepsilon_1-\varepsilon_0}{\hbar\omega_1}, \quad |\zeta|=\frac{64}{81}e_0cZ\alpha\frac{\bar{E}}{\omega_1(\varepsilon_1-\varepsilon_0)}. \tag{10}$$

The quantity $2\hbar\omega_1\kappa$, proportional to the intensity of the electromagnetic wave, is the relative shift of the energy of the atomic levels in the electromagnetic field. In a sufficiently strong field, this is a rather appreciable quantity. Thus, for the hydrogen atom at $\omega_1 = 10^{14}$ sec^{-1} and $\bar{E} = 10^6$ V/cm we have $\kappa \approx 0.6$ ($\zeta \approx 0.17$), i.e., the energy level shift coincides in order of magnitude with the energy of the laser quantum. In view of such a large energy shift, the frequencies of absorption of the light by the Hamiltonian differ greatly in the presence and in the absence of a strong field. The formula obtained by us for the energy shift agrees with the results of Ritus[3].

We note that calculation of the probability of the absorption of light by the atom, carried out on the basis of the nonrelativistic Hamiltonian

$$H'_{int}=\frac{e_0\mathbf{pA}(t)}{m}+\frac{e_0^2\mathbf{A}^2(t)}{2m},$$

leads to a formula that coincides exactly with formula (9). As shown by this calculation, that part of the operator H'_{int} which is quadratic in the vector potential makes no contribution to the transition probability (if the inequality (7) is satisfied).

An attempt to solve the problem of the absorption of weak electromagnetic radiation by a hydrogen atom in the presence of a strong electromagnetic wave was undertaken earlier by Kovarskiĭ[8]. The formula obtained by us (9) for the absorption probability differs significantly from the corresponding formula given in[8], both in the arguments of the Bessel functions and in the energy-conservation laws. Kovarskiĭ's results[8] seem to be in error[1)] because he did not take into account in the initial Hamiltonian the matrix element

$$d_{1s,2p}=\int d\mathbf{r}\varphi_{1s}^*(\mathbf{r})ez\varphi_{2p}(\mathbf{r})$$

(Allowance for this transition matrix element leads, in particular, to a shift of the atomic energy levels in an electromagnetic field).

1)Erratum (published in Zh. Eksp. Teor. Fiz. 68, No. 2, February 1975). The matrix element $d_{1s,2p}$ referred to in our paper was taken into account in a paper by V. A. Kovarskiĭ (Zh. Eksp. Teor. Fiz. 57, 1613 (1969)–Sov. Phys.–JETP 30, 872 (1970)), of which the authors, unfortunately, unaware at the time the present paper was written. As correctly pointed out to us by V. A. Kovarskiĭ, to obtain the correct numerical value of the main parameter κ of the theory it is necessary to take into account the entire energy spectrum of the hydrogen atom, as was indeed done in his article. The fact that we used a two-level model of the atom, which took into account only the 1s, 2s, and 2p states, has caused the parameter κ to be overestimated by many times.

1L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys.-JETP **20**, 1307 (1965)].
2V. M. Buĭmistrov and V. P. Oleĭnik, Fiz. Tekh. Poluprovodn. 1, 85 (1967) [Sov. Phys.-Semicond. 1, 65 (1967)].
3V. I. Ritus, Zh. Eksp. Teor. Fiz. **51**, 1544 (1966) [Sov. Phys.-JETP **24**, 1041 (1967)].
4V. P. Oleĭnik, Zh. Eksp. Teor. Fiz. **61**, 27 (1971) [Sov. Phys.-JETP **34**, 14 (1972)].
5V. P. Oleĭnik, Zh. Eksp. Teor. Fiz. **53**, 1997 (1967) [Sov. Phys.-JETP **26**, 1132 (1968)]; ZhETF Pis. Red. 9, 530 (1969) [JETP Lett. 9, 321 (1969)].
6A. I. Akhiezer and V. B. Berestetskiĭ, Kvantovaya élektrodinamika (Quantum Electrodynamics), Nauka (1969).
7V. V. Tolmachev, Teoriya fermi-gaza (Theory of Fermi Gas) Moscow State Univ. (1973), pp. 214, 215.
8V. A. Kovarskiĭ, Zh. Eksp. Teor. Fiz. **57**, 1217 (1969) [Sov. Phys.-JETP **30**, 663 (1970)].

Translated by J. G. Adashko.
105