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MULTIPHOTON TRANSITIONS IN THE DISCRETE SPECTRUM OF ATOMS AND IONIZATION PROCESSES IN A STRONG ELECTRIC FIELD

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Multiphoton transitions in the discrete spectrum of atoms and multiphoton ionization processes, caused by a change in the frequency spectrum of the photons due to the electron-photon interaction, are considered. The problem of the absorption of light of frequency Ω by an atom in the presence of nonresonance laser radiation of frequency $\omega \ll \Omega$ is solved. An expression is obtained for the probability of multiphoton ionization of an atom, and its dependence on the statistical properties of the radiation is investigated.

THE idea of the possibility of many-quantum processes associated with the transition of an electron from one energy state to another because of a change of the frequencies of the Bose (phonon) subsystem which interacts with the electron, was first expressed in well-known articles by Ya. I. Frenkel'.[1] The inclusion of part of the interaction in the zero-order approximation for the Hamiltonian is achieved at the expense of the adiabatic approximation. The mathematical basis which guarantees the many-quantum character of the transition consists in the fact that it is impossible to diagonalize, by a single unitary transformation, two quadratic forms in the boson amplitudes which refer to different electron states.[2]

In the case of the electron-photon system, the utilization of the method of successive diagonalization of the Hamiltonian by unitary transformation is a convenient scheme of calculation. Both multiphoton transitions in the discrete spectrum of atoms and the problem of multiphoton ionization will be investigated below. In the latter case, in contrast to articles[3-6] basic attention will be given to the dependence of the probability for an ionization process on the statistical properties of the incident electromagnetic radiation.

1. TRANSFORMATION OF THE HAMILTONIAN OF THE ELECTRON-PHOTON SYSTEM

Let us consider the Hamiltonian of the electron-photon system for a hydrogen-like atom interacting with radiation (the case of the l-degeneracy of hydrogen is investigated separately[7]):

$$\hat{H} = \hat{H}^0 + \hat{H}' + \hat{H}'', \tag{1}$$

$$\hat{H}^0 = \sum_i \varepsilon_i a_i^+ a_i + \sum_{\varkappa\lambda} \hbar c \varkappa (b_{\varkappa\lambda}^+ b_{\varkappa\lambda} + 1/2), \tag{2}$$

$$\hat{H}' = \varepsilon' \sum_{ij\varkappa\lambda} a_i^+ a_j [V_{ij\varkappa\lambda} b_{\varkappa\lambda} + V_{ji\varkappa\lambda} b_{\varkappa\lambda}^+],$$

$$\hat{H}'' = \varepsilon'' \sum_{ij\varkappa_1\lambda_1\varkappa_2\lambda_2} a_i^+ a_j \{A^{(ij)}_{\varkappa_1\lambda_1\varkappa_2\lambda_2} b_{\varkappa_1\lambda_1} b_{\varkappa_2\lambda_2} + A^{(ij)}_{\varkappa_1\lambda_1,-\varkappa_2\lambda_2} b_{\varkappa_1\lambda_1} b^+_{\varkappa_2\lambda_2} \tag{3}$$

$$+ A^{(ij)*}_{\varkappa_1\lambda_1\varkappa_2\lambda_2} b^+_{\varkappa_1\lambda_1} b^+_{\varkappa_2\lambda_2} + A^{(ij)*}_{\varkappa_1\lambda_1,-\varkappa_2\lambda_2} b^+_{\varkappa_1\lambda_1} b_{\varkappa_2\lambda_2}\}, \tag{4}$$

$$a_i^+ a_j + a_j a_i^+ = \delta_{ij}, \quad b_{\varkappa\lambda} b^+_{\varkappa_1\lambda_1} - b^+_{\varkappa_1\lambda_1} b_{\varkappa\lambda} = \delta_{\varkappa_1\varkappa} \delta_{\lambda_1\lambda}, \tag{5}$$

$$V_{ij\varkappa\lambda} = \frac{i\hbar e_0}{mc} \sqrt{\frac{2\pi\hbar c}{|\varkappa|}} L^{-3/2} \int \psi_i^*(r) e^{i\varkappa r} \mathbf{e}_{\varkappa\lambda} \nabla \psi_j(r)\, dv, \tag{6}$$

$$A^{(ij)}_{\varkappa_1\lambda_1,\pm\varkappa_2\lambda_2} = \frac{\pi e_0^2 \hbar}{mcL^3} \frac{\mathbf{e}_{\varkappa_1\lambda_1} \mathbf{e}_{\varkappa_2\lambda_2}}{\sqrt{|\varkappa_1||\varkappa_2|}} \int \psi_i^*(r) \psi_j(r) e^{i(\varkappa_1 \pm \varkappa_2) \mathbf{r}}\, dv \tag{7}$$

(e_0, m, and c respectively denote the electron's charge, its mass, and the velocity of light). In formulas (3)—(7) the following notation is used: ϵ_i and ψ_i are the energy eigenvalue and the wave function of the atom in state i (i includes the necessary set of quantum numbers); a_i^+ and a_i are Fermi operators for the creation and annihilation of electrons; $b_{\kappa\lambda}^+$ and $b_{\kappa\lambda}$ are Bose operators for a photon with wave vector κ and polarization λ (λ takes two values; $e_{\kappa\lambda}$ is the unit polarization vector); L^3 is the volume of quantization; ϵ' and ϵ'' are formal parameters of smallness[1)] ($\epsilon' \equiv 1$, $\epsilon'' \equiv 1$).

Let us use the method of successive diagonalization of the Hamiltonian by unitary transformation:[8]

$$\tilde{H} = e^{-\hat{S}} \hat{H} e^{\hat{S}} = \hat{H} + [\hat{H}\hat{S}] + \frac{1}{2!}[[\hat{H}\hat{S}]\hat{S}] + \ldots,$$

$$[\hat{A}\hat{B}] \equiv \hat{A}\hat{B} - \hat{B}\hat{A}.$$

The operator $\hat{S}$ should be chosen from the condition that the terms of first order ($\sim \epsilon'$) should vanish. Since the term H″ is much smaller than the term $\hat{H}'$, it may be assigned to the terms of second order of smallness: $\epsilon'' \sim (\epsilon')^2$. We obtain

$$\hat{H}' + [\hat{H}^0 \hat{S}] = 0,$$

$$\hat{S} = \varepsilon' \sum_{ij\varkappa\lambda} \left\{ \frac{V_{ij\varkappa\lambda}}{\varepsilon_j - \varepsilon_i + \hbar\omega_\varkappa} b_{\varkappa\lambda} + \frac{V^*_{ji\varkappa\lambda}}{\varepsilon_j - \varepsilon_i - \hbar\omega_\varkappa} b^+_{\varkappa\lambda} \right\} a_i^+ a_j. \tag{8}$$

The transformed Hamiltonian has the form

$$\tilde{H} = \hat{H}^0 + \hat{H}'' + \hat{W}' + \hat{W}''.$$

Here the following notation is used:

$$\hat{W}' = {}^1/_2 [\hat{H}'\hat{S}], \quad \hat{W}'' = {}^1/_2 [\hat{S}[\hat{S}\hat{H}']] + [\hat{H}'\hat{S}] + \ldots. \tag{9}$$

It is convenient to split the operators $\hat{W}'$ and $\hat{W}''$ into parts $\hat{W}_d'$ and $\hat{W}_d''$ which are diagonal with respect to the Fermi amplitudes, and nondiagonal parts $\hat{W}_{nd}'$ and $\hat{W}_{nd}''$. In this connection

$$\langle i|\hat{W}'_d|i\rangle \neq 0, \quad \langle i|\hat{W}'_{nd}|i\rangle = 0, \quad \langle i|\hat{W}''_d|i\rangle \neq 0, \quad \langle i|\hat{W}''_{nd}|i\rangle = 0.$$

The operators $\hat{W}_{nd}'$ and $\hat{W}_{nd}''$ mix different electron states and may be regarded as perturbations which

1)The introduction of these parameters corresponds to the usual approach of quantum electrodynamics.

generate multi-quantum ionization. (For simplicity we shall omit the small "anharmonic" term $\hat{W}_d''^{2)}$.)

Let us write out the Hamiltonian for the photon subsystem in the i-th electron state:

$$\tilde{H}_i \equiv \langle i|\tilde{H}|i\rangle = \tilde{\varepsilon}_i + \sum_{\varkappa_1\lambda_1\varkappa_2\lambda_2} \{{}^1/_2 B^{(i)}_{\varkappa_1\lambda_1\varkappa_2\lambda_2} b_{\varkappa_1\lambda_1} b_{\varkappa_2\lambda_2} + {}^1/_2 B^{(i)*}_{\varkappa_1\lambda_1\varkappa_2\lambda_2} b^+_{\varkappa_1\lambda_1} b^+_{\varkappa_2\lambda_2} + [\hbar\omega_{\varkappa_1}\delta_{\varkappa_1\varkappa_2}\delta_{\lambda_1\lambda_2} + D^{(i)}_{\varkappa_1\lambda_1\varkappa_2\lambda_2}] b^+_{\varkappa_1\lambda_1} b_{\varkappa_2\lambda_2}\}, \quad (10)$$

$$\tilde{\varepsilon}_i \equiv \varepsilon_i + (\varepsilon')^2 \sum_{j\varkappa\lambda} \frac{|V_{ij\varkappa\lambda}|^2}{\varepsilon_i - \varepsilon_j - \hbar\omega_\varkappa} + \varepsilon'' \sum_{\varkappa\lambda} A^{(ii)}_{\varkappa\lambda;\,-\varkappa\lambda}.$$

Here

$$B^{(i)}_{\varkappa_1\lambda_1\varkappa_2\lambda_2} = \frac{(\varepsilon')^2}{2}\left\{\sum_j \left(\frac{V_{ij\varkappa_1\lambda_1}V_{ji\varkappa_2\lambda_2}}{\varepsilon_i - \varepsilon_j + \hbar\omega_{\varkappa_2}} + \frac{V_{ij\varkappa_2\lambda_2}V_{ji\varkappa_1\lambda_1}}{\varepsilon_i - \varepsilon_j + \hbar\omega_{\varkappa_1}}\right) + \sum_j \left(\frac{V_{ji\varkappa_1\lambda_1}V_{ij\varkappa_2\lambda_2}}{\varepsilon_i - \varepsilon_j - \hbar\omega_{\varkappa_2}} + \frac{V_{ji\varkappa_2\lambda_2}V_{ij\varkappa_1\lambda_1}}{\varepsilon_i - \varepsilon_j - \hbar\omega_{\varkappa_1}}\right)\right\} + 2\varepsilon'' A^{(ii)}_{\varkappa_1\lambda_1\varkappa_2\lambda_2}, \quad (11)$$

$$D^{(i)}_{\varkappa_1\lambda_1\varkappa_2\lambda_2} = (\varepsilon')^2 \sum_j \left\{\frac{V^*_{ji\varkappa_1\lambda_1}V_{ji\varkappa_2\lambda_2}}{\varepsilon_i - \varepsilon_j + \hbar\omega_{\varkappa_2}} + \frac{V^*_{ij\varkappa_1\lambda_1}V_{ij\varkappa_2\lambda_2}}{\varepsilon_i - \varepsilon_j - \hbar\omega_{\varkappa_2}}\right\} + \varepsilon''[A^{(ii)*}_{\varkappa_1\lambda_1,\,-\varkappa_2\lambda_2} + A^{(ii)}_{\varkappa_2\lambda_2,\,-\varkappa_1\lambda_1}]. \quad (12)$$

The coefficients of the quadratic form satisfy the following relations:

$$D^{(i)}_{\varkappa_1\lambda_1\varkappa_2\lambda_2} = D^{(i)*}_{\varkappa_2\lambda_2\varkappa_1\lambda_1}, \quad B^{(i)}_{\varkappa_1\lambda_1\varkappa_2\lambda_2} = B^{(i)}_{\varkappa_2\lambda_2\varkappa_1\lambda_1}.$$

For simplicity, we shall investigate the multiphoton transitions of an electron from the ground 1s-state of the atom. To within terms $\sim(\epsilon')^2$ and ϵ'', the initial Hamiltonian $\tilde{H}_s$ can be diagonalized[9] with the aid of a canonical transformation to new variables $\xi^+_{\kappa\lambda}$ and $\xi_{\kappa\lambda}$. We find:

$$\tilde{H}_s = J_s + \sum_{\varkappa\lambda} \hbar\tilde{\omega}_{\varkappa\lambda}\xi^+_{\varkappa\lambda}\xi_{\varkappa\lambda},$$

$$J_s = \varepsilon_s - \sum_{\varkappa\lambda\varkappa_1\lambda_1} \frac{\omega_{\varkappa_1}}{\hbar}\left[\frac{B^{(s)}_{\varkappa\lambda\varkappa_1\lambda_1}}{\omega_\varkappa + \omega_{\varkappa_1}}\right]^2, \quad \tilde{\omega}_{\varkappa\lambda} = \omega_\varkappa + \frac{1}{\hbar} D^{(s)}_{\varkappa\lambda\varkappa\lambda}. \quad (13)$$

The photon Hamiltonian $\tilde{H}_p$ corresponding to the p-th excited state of the electron is determined by formula (10). Let us transform it to the variables $\xi^+_{\kappa\lambda}$ and $\xi_{\kappa\lambda}$ and retain the terms $\sim(\epsilon')^2$ and ϵ''. We obtain

$$\tilde{H}_p = J_p + \sum_{\varkappa\lambda} \hbar\tilde{\omega}_{\varkappa\lambda}\xi^+_{\varkappa\lambda}\xi_{\varkappa\lambda} + \sum_{\varkappa_1\lambda_1\varkappa_2\lambda_2} \{{}^1/_2 B^{(ps)*}_{\varkappa_1\lambda_1\varkappa_2\lambda_2}\xi^+_{\varkappa_1\lambda_1}\xi^+_{\varkappa_2\lambda_2} + {}^1/_2 B^{(ps)}_{\varkappa_1\lambda_1\varkappa_2\lambda_2}\xi_{\varkappa_1\lambda_1}\xi_{\varkappa_2\lambda_2} + D^{(ps)}_{\varkappa_1\lambda_1\varkappa_2\lambda_2}\xi^+_{\varkappa_1\lambda_1}\xi_{\varkappa_2\lambda_2}\}. \quad (14)$$

Here

$$J_p = \tilde{\varepsilon}_p - \sum_{\varkappa_1\lambda_1\varkappa_2\lambda_2} \frac{\omega_{\varkappa_1}}{\hbar}\left[\frac{B_{\varkappa_1\lambda_1\varkappa_2\lambda_2}}{\omega_{\varkappa_1} + \omega_{\varkappa_2}}\right]^2,$$

$$B^{(ps)}_{\varkappa_1\lambda_1\varkappa_2\lambda_2} \equiv B^{(p)}_{\varkappa_1\lambda_1\varkappa_2\lambda_2} - B^{(s)}_{\varkappa_1\lambda_1\varkappa_2\lambda_2},$$

$$D^{(ps)}_{\varkappa_1\lambda_1\varkappa_2\lambda_2} \equiv D^{(p)}_{\varkappa_1\lambda_1\varkappa_2\lambda_2} - D^{(s)}_{\varkappa_1\lambda_1\varkappa_2\lambda_2}. \quad (15)$$

2. MULTIPHOTON SPECTROSCOPY OF ATOMS

Let us consider multiphoton transitions in the discrete spectrum of atoms associated with the interaction with steady-state laser radiation of a given frequency ω, intensity $\mathscr{L}(\omega)$ and polarization λ_0. Since the resonance condition for a given electron transition and a fixed frequency is usually not satisfied, it is convenient to introduce an additional source of radiation with an adjustable frequency Ω of generation. In this connection the law of energy conservation for the absorption of laser photons and of the photon Ω is always satisfied owing to the appropriate choice of Ω. Let us denote the spectral intensity of the light Ω by $\mathscr{L}(\Omega)$, and for definiteness let us assume z-polarization for it.

The probability dW of absorption per unit time of a quantum of frequency Ω, lying in the interval between Ω and $\Omega + d\Omega$, is determined by the Fourier transform of the correlation function K_Ω of the dipole moment operators $\hat{d}$:[10]

$$dW = \frac{4\pi^2}{\hbar^2 c}\mathscr{L}(\Omega)\mathscr{K}_\Omega\, d\Omega, \quad \mathscr{K}_\Omega = \frac{1}{2\pi}\int_{-\infty}^{+\infty}\mathscr{K}(t)e^{-i\Omega t}\,dt,$$

$$\mathscr{K}(t) = \mathrm{Av}\{\hat{d}(0)\hat{d}(t)\}, \quad \hat{d}(t) = \exp\left\{\frac{i\hat{H}t}{\hbar}\right\}\hat{d}\exp\left\{-\frac{i\hat{H}t}{\hbar}\right\}; \quad (16)$$

Av{...} denotes the operation of statistical averaging. Let the frequency Ω be chosen so that a resonance electron transition from the ground state s to a given discrete level p is realized. The formula for $\mathscr{K}(t)$ can be transformed to the form[2]

$$\mathscr{K}(t) = d_{sp}{}^2\,\mathrm{Sp}\,\{\exp(i\tilde{H}_p t/\hbar)\exp(-i\tilde{H}_s t/\hbar)\rho_F\}. \quad (17)$$

Here d_{sp} is the matrix element of the z-component of the dipole moment operator, $\tilde{H}_p$ and $\tilde{H}_s$ are determined by formulas (14) and (13),[3)] and ρ_F is the statistical operator for the electromagnetic field of the laser radiation.

Let us introduce ordering into formula (17) with the aid of a change to the T-ordered exponential:

$$\mathscr{K}(t) = d_{sp}{}^2\,\mathrm{Sp}\left\{\rho_F T\exp\left(-\frac{i}{\hbar}\int_0^t V(t_1)\,dt_1\right)\right\}, \quad (18)$$

$$V(t_1) = \exp(i\tilde{H}_s t_1/\hbar)(\tilde{H}_p - \tilde{H}_s)\exp(-i\tilde{H}_s t_1/\hbar). \quad (19)$$

In order to evaluate the average in formula (18), we shall use the $\mathscr{P}$-representation of the density operator ρ_F considered by Glauber[11]

$$\rho_F = \int \mathscr{P}(\{\beta_\varkappa\})\prod_\varkappa |\beta_\varkappa\rangle\langle\beta_\varkappa|\,d^2\beta_\varkappa. \quad (20)$$

Here the β_κ denote the eigenvalues of the annihilation operator ξ_κ for the mode κ:

$$\xi_\varkappa|\beta_\varkappa\rangle = \beta_\varkappa|\beta_\varkappa\rangle,$$

$\mathscr{P}(\{\beta_\kappa\})$ is a weight function whose form is determined by Glauber[11] for certain models of a light beam. In the steady-state case $P \equiv P(\{|\beta_\kappa|\})$, so that if $\beta_\kappa = r_\kappa e^{i\theta_\kappa}$ then integration over the phases of the amplitudes is not directly related to the form of the function $\mathscr{P}$.

Below we confine our attention to a single-mode approximation for a laser beam propagating in the direction $\kappa = \kappa_0$ and having a polarization λ_0.

Let us carry out the operation of averaging in formula (18) with the aid of the density operator (20). As a preliminary it is necessary to expand the T-ordered exponential appearing in Eq. (18) in a series. In the case of sufficiently intense radiation, the average number of photons in the mode is large and one can neglect the noncommutativity of the field operators. Upon aver-

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[2)]This approximation (for a two-level model) limits the admissible value of the intensity F of the light wave's field by the condition (x_{12} denotes the matrix element of the coordinate): $|e_0{}^2x_{12}{}^2F^2/\hbar^2(\omega_{12}{}^2 - \omega^2)| < 1$. Additional restrictions due to the presence of a continuous spectrum do not arise since the matrix element $\langle k\,|\,\hat{W}''_d\,|\,k\rangle$ is rigorously equal to zero.

[3)]It is obvious that $J_s \approx \epsilon_s$, $J_p \approx \epsilon_p$.

aging, due to the integration over the phases, only those terms of the series survive for which the number of creation operators is equal to the number of annihilation operators. We find:

$$\mathscr{K}(t) = d_{sp}^2 \sum_{l_1=0}^{\infty} \sum_{l_2=0}^{\infty} \frac{(-1)^{l_2}}{l_1!(l_2!)^2} \left[\frac{it}{\hbar} D^{(ps)}_{\varkappa_0\lambda_0\varkappa_0\lambda_0}\right]^{l_1} \left[\frac{B^{(ps)}_{\varkappa_0\lambda_0\varkappa_0\lambda_0} \sin \widetilde{\omega}_{\varkappa_0\lambda_0} t}{2\hbar\widetilde{\omega}_{\varkappa_0\lambda_0}}\right]^{2l_2} \varphi_{l_1 l_2}, \quad (21)$$

where

$$\varphi_{l_1 l_2} = \int \ldots \int \mathscr{P}(\{r_\varkappa\}) r_\varkappa^{4l_2+2l_1} \prod_\varkappa d^2\beta_\varkappa. \quad (22)$$

For a stably oscillating laser the weight function which describes the radiation has a δ-shape:[11]

$$\mathscr{P}(r_\varkappa) = \frac{1}{2\pi\sqrt{\bar{n}_{\varkappa_0\lambda_0}}} \delta(|\beta_{\varkappa_0}| - \sqrt{\bar{n}_{\varkappa_0\lambda_0}}). \quad (23)$$

Here $\bar{n}_{\kappa_0\lambda_0}$ is the average number of photons in the mode. With the aid of Eq. (23) we find

$$\varphi_{l_1 l_2} = (\bar{n}_{\varkappa_0\lambda_0})^{2l_2+l_1}. \quad (24)$$

Let us perform the necessary summation in formula (21). Let us utilize the dipole approximation for the constants of the theory. Finally we obtain

$$\mathscr{K}(t) = d_{sp}^2 \exp\{it(\Omega_{ps} + 2\rho_1\omega)\} J_0(2\rho_1 \sin\omega t), \quad (25)$$

$$\Omega_{ps} \equiv \frac{\varepsilon_p - \varepsilon_s}{\hbar}, \quad 2\rho \equiv D^{(ps)}_{\varkappa_0\lambda_0\varkappa_0\lambda_0}\bar{n}_{\varkappa_0\lambda_0} = B^{(ps)}_{\varkappa_0\lambda_0\varkappa_0\lambda_0}\bar{n}_{\varkappa_0\lambda_0}, \quad (26)$$

$\rho_1 = \rho/\hbar\omega$, and $J_0(x)$ is the Bessel function of real argument. The constant ρ may be expressed in a simple way in terms of the matrix elements of the coordinates:[12] $\rho = \rho_p - \rho_s$, where ρ_p and ρ_s are given by the formula ($i = s, p$; e_i—unit polarization vector)

$$\rho_i = 2\pi e_0^2 \hbar\omega \bar{n}_{\varkappa_0\lambda_0} L^{-3} \sum_j \frac{(\varepsilon_i - \varepsilon_j)\langle i|\mathbf{r}e_i|j\rangle\langle j|\mathbf{r}e_i|i\rangle}{(\varepsilon_i - \varepsilon_j)^2 - \hbar^2\omega^2} \quad (27)$$

To within a factor, the structure of ρ_i coincides with the Stark shift of the level in a high-frequency field.[13]

Let us determine dW_{sp}. We perform the necessary integration over t by using the expansion

$$J_0(2\rho_1 \sin\omega t) = J_0^2(\rho_1) + 2\sum_{n=1}^{\infty} J_n^2(\rho_1)\cos 2n\,\omega t,$$

where $J_n(\rho_1)$ denotes a Bessel function. We find

$$dW_{sp} = \frac{4\pi^2}{\hbar^2 c} d_{sp}^2 \mathscr{L}(\Omega) \sum_{n=0}^{\infty} J_n^2(\rho_1)\,\delta(\Omega_{ps} + 2\rho_1\omega - \Omega - 2n\omega)\, d\Omega. \quad (28)$$

As follows from formula (28), the probability dW_{sp} has the characteristic form of a sum of terms which describe multiphoton transitions.

The presence of the term $2\rho_1\omega$ in the law of energy conservation leads to a shift of the maximum for absorption of the frequency Ω to the "blue" side. For example, if the frequency Ω was tuned to the resonance $\Omega = \Omega_{ps}$, then the switching-on of sufficiently intense laser radiation "destroys" the resonance. Here the term $2\rho_1\omega$, with regard to its own magnitude, must at least exceed the width of the absorption line. The integral probability of a multiphoton transition in the discrete spectrum of an atom is determined by the formula

$$W_{ss} \equiv \int dW_{sp} = \frac{4\pi^2}{\hbar^2 c} d_{sp}^2 \mathscr{L}(\Omega) J^2_{[n]}(\rho_1). \quad (29)$$

Here $[n]$ denotes the integer part of n,

$$2n\omega = \Omega_{ps} + 2\rho_1\omega - \Omega.$$

For $\rho_1 = 0$ expression (29) goes over into the formula for the probability $W_{sp}^{(1)}$ of the single-photon absorption of light. For $\rho_1 \neq 0$, $\omega \ll \Omega$ we have $\Omega = \Omega_{ps} + 2\rho_1\omega$, and

$$W_{sp} = W_{sp}^{(1)} J_0^2(\rho_1). \quad (30)$$

The presence of the square of the Bessel function in Eq. (30) is typical and reflects the contribution of virtual laser photons to the transition.[8,13] In the general case of an n-photon transition its probability is entirely determined by the factor $J^2_{[n]}(\rho_1)$. In the case of only a single laser source, it is most probable that one of the strongly excited levels of the atom will satisfy the resonance condition. In the presence of a transition to such a level, photoionization of the atom will subsequently occur with a probability close to unity. Since the "bottleneck" of the process is the multiphoton excitation to this discrete level, then the corresponding probability is simultaneously the probability of multiphoton ionization of the atom. In principle indirect ionization may lead to smaller values of the "index of the multiphoton nature" n in the law $W_{\text{ioniz}} \sim F^{2n}$ (F is the field intensity of the laser beam) and must be taken into account side by side with other possible physical considerations.[14-16]

3. DIRECT MULTIPHOTON IONIZATION OF ATOMS

Let us consider multiphoton ionization of an atom from the ground state due to the influence of the perturbation $\hat{W}_{nd}$:

$$\hat{W}_{nd} = \hat{W}'_{nd} + \hat{W}''_{nd}.$$

After standard transformations the expression for the transition probability per unit time may be reduced to the form

$$W_{\text{ioniz}} = \mathrm{Sp}\left\{\frac{\rho_F}{\hbar}\sum_{\mathbf{k}}\int_{-\infty}^{+\infty} \langle s|\hat{W}_{nd}|\mathbf{k}\rangle\, T\exp\left\{-\frac{i}{\hbar}\int_0^t \hat{V}(t_1)dt_1\right\}\langle \mathbf{k}|\hat{W}_{nd}(t)|s\rangle\, dt\right\}.$$

Here $\hat{V}(t)$ is the same operator as in Eq. (19), but the index p = k refers to a state of the continuous spectrum,

$$\hat{W}_{nd}(t) = \exp\{i\hat{H}_s t/\hbar\}\hat{W}_{nd}\exp\{-i\hat{H}_s t/\hbar\}.$$

The following calculation does not differ fundamentally from the calculation for the case of a discrete spectrum.

In the case of a stably oscillating laser (L) we find

$$W^{(L)}_{\text{ioniz}} = W^{0(L)}_{\text{ioniz}} + \Delta W, \quad (31)$$

$$W^{0(L)}_{\text{ioniz}} = \sum_{n_1=n_0}^{\infty} W_2(n_1;\omega) J_{n_1}^2(\rho_1) + \sum_{m_1=m_0}^{\infty} W_3(m_1;\omega) J_{m_1}^2(\rho_1). \quad (32)$$

Here $J_k(x)$ denotes a Bessel function; the contribution to the transition probability from the terms W_{nd} which do not enter into $W_2(n_1;\omega)$ and $W_3(m_1;\omega)$ (see formula (39) below) is denoted by ΔW

$$W_2(n_1;\omega) = \frac{mk_\omega}{4\pi^2\hbar^3} N^2_{\varkappa_0\lambda_0} L^9 \int d\Omega_{\mathbf{k}_\omega} \times \left|\sum_{j_1} \frac{V_{sj_1}V_{j_1k_\omega}(\varepsilon_{k_\omega} - \varepsilon_{j_1} - \hbar\omega - n_1\hbar\omega + \rho_1\hbar\omega)}{(\varepsilon_s - \varepsilon_{j_1} + \hbar\omega)(\varepsilon_{k_\omega} - \varepsilon_{j_1} - \hbar\omega)}\right|^2, \quad (33)$$

$$W_3(m_1;\omega) = \frac{mk'_\omega N^3_{\varkappa_0\lambda_0} L^{12}}{8\pi^2\hbar^3}\int d\Omega_{\mathbf{k}_\omega},$$

$$\times\left|\sum_{j_1 j_2}\frac{V_{sj_1}V_{j_1j_2}V_{j_2k_\omega}\{3(\varepsilon_{j_1}-\varepsilon_{j_2}+\hbar\omega)+2m_1\hbar\omega-2\rho_1\hbar\omega\}}{2(\varepsilon_s-\varepsilon_{j_1}+\hbar\omega)(\varepsilon_{j_1}-\varepsilon_{j_2}+\hbar\omega)(\varepsilon_s-\varepsilon_{j_2}+2\hbar\omega+2m_1\hbar\omega+2\rho_1\hbar\omega)}\right|^2, \quad (34)$$

$$q=\frac{|\varepsilon_s|}{\hbar\omega}+2\rho_1,\ k_\omega=\sqrt{\frac{2m\omega}{\hbar}(2n-q+2)},\ \ k_\omega'=\sqrt{\frac{2m\omega}{\hbar}(2n-q+3)}. \quad (35)$$

Since the Bessel functions decrease sharply with increasing values of n_1 and m_1, one can restrict oneself to the formula

$$\hat{W}^{o(L)}_{\text{ioniz}}=W_2(\omega)J_{n_0}^2(\rho_1)+W_3(\omega)J_{m_0}^2(\rho_1), \quad (36)$$

$$n_0=\frac{1}{2}\begin{cases}[q-1], & \text{if } [q-2] \text{ is odd}\\ [q], & \text{if } [q-2] \text{ is even}\end{cases}$$

$$m_0=\frac{1}{2}\begin{cases}[q-2], & \text{if } [q-3] \text{ is odd}\\ [q-1], & \text{if } [q-3] \text{ is even}\end{cases} \quad (37)$$

Here $W_2(\omega) \equiv W_2(n_0;\omega)$ and $W_3(\omega) \equiv W_3(m_0;\omega)$. As is not difficult to verify, in the case of two- and three-photon processes the quantities $W_2(\omega)$ and $W_3(\omega)$ correspond to the probabilities of two-photon and three-photon ionization of the atom. For multiphoton transitions of arbitrary order, these quantities must be numerically estimated in each specific case. The basic parameter of the theory, $\rho = \rho_{k_\omega} - \rho_s$ is calculated according to formula (27). For the calculation of ρ_s one can assume $\hbar\omega \ll |\epsilon_s|$. In connection with a calculation of ρ_{k_ω}, the excited states primarily give a contribution to the sum over j in formula (27). In the approximation $\hbar\omega \gg |\epsilon_{k_\omega} - \epsilon_j|$ the evaluation of ρ_{k_ω} can be carried out exactly on the basis of a sum rule (in analogy to the calculation of the Thomson cross-section for the scattering of light[17]). For a sufficiently large ionization potential ($\rho_{k_\omega} \gg \rho_s$) we find

$$\rho_1\approx\frac{\rho_{k_\omega}}{\hbar\omega}\approx-\frac{2\pi e_0^2\bar{n}_{\varkappa_0\lambda_0}L^{-3}}{\hbar^2\omega^2}\sum_j(\varepsilon_k-\varepsilon_j)\langle k|\mathbf{re}_k|j\rangle\langle j|\mathbf{re}_k|k\rangle,$$

$$\rho_1=\frac{\pi e_0^2}{m\omega^2}N_{\varkappa_0\lambda_0}\equiv\frac{|\varepsilon_s|}{2\hbar\omega\gamma^2},\quad \gamma=\frac{\omega}{\omega_t}, \quad (38)$$

where ω_t is the tunneling frequency.[3]

Let us estimate the magnitude of ΔW. First of all we note that since the contribution of the terms H'' ($H'' \sim A^2$, A is the vector potential) to the probability of two- and three-photon transitions is, according to numerous estimates, less important than the contribution from the term $\mathbf{A}\cdot\nabla$, then for the sake of simplicity it is not taken into consideration in formulas (33) and (34) for $W_2(\omega)$ and $W_3(\omega)$. The structure of ΔW is approximately represented by a sum of the form

$$\Delta W \sim W_4(\omega)J_{p_0}^2(\rho_1)+W_5(\omega)J_{l_0}^2(\rho_1)+\ldots \quad (39)$$

(p_0 and l_0 satisfy conditions of the type (37); for example, if $[q-4]$ is even then $p_0 = (1/2)[q-2]$ and so forth). The coefficients $W_4(\omega)$, $W_5(\omega)$ and so on correspond to the probabilities of four-, five-, and so on photon transitions under the assumption that the frequency ω satisfies a given multiphoton resonance.

It is not difficult to verify that the contribution ΔW is small in comparison with $W^{o(L)}_{\text{ioniz}}$. For example, in the case of four-photon ionization

$$W^{o(L)}_{\text{ioniz}}=W_2(\omega)J_1^2(\rho_1),\quad \Delta W=W_4(\omega)J_2^2(\rho_1)$$

and an estimate shows that ΔW is approximately an order of magnitude smaller than $W^{o(L)}_{\text{ioniz}}$. One can understand the last result if one takes into consideration that in the case of an n-photon transition $W_n(\omega)$ decreases like $[(n-1)!]^{-2}$ while the square of the Bessel function which enters into $W^{o(L)}_{\text{ioniz}}$ (for $\rho_1 < 1$ and $n \gg 1$) falls off like $2^{-(n-2)}[(n-2/2)!]^{-2}$. (The remaining factors are approximately identical for comparable terms). We neglect the contribution of ΔW in what follows.

It is not difficult to verify that in the limiting cases formula (36) goes over into the well-known expressions for the ionization probability. For $\rho_1 \ll 1$ and a large number of photons we find

$$W^{o(L)}_{\text{ioniz}}=W_2^0\left(\frac{2{,}718e_0^2F^2}{8\hbar\omega^3m[q+1]}\right)^{2n_0+1}+W_3^0\left(\frac{2{,}718e_0^2F^2}{8\hbar\omega^3m[q+1]}\right)^{2m_0+2} \quad (40)$$

The constants W_2^0 and W_3^0 correspond to the quantities $W_2(\omega)$ and $W_3(\omega)$ taken at a value for the photon density $N^0 \approx m\omega^2\sqrt{[q]}/\pi e_0^2$, and F denotes the amplitude of the light wave's field intensity. The expression standing inside the circular brackets in formula (40) determines the "threshold" dependence of the transition probability and agrees with the analogous expression in the work by L. V. Keldysh[3] (formula (21)). (An estimate of W_2^0 for the hydrogen atom gives: $W_2^0 \sim 10^{17}\ \text{sec}^{-1}$). In another limiting case, for $\rho_1 \gg 1$, we obtain

$$J_{n_0}^2(\rho_1)\approx\frac{q_0}{3\pi^2\rho_1}\left[K_{1/3}\left\{\frac{q_0^{3/2}}{3\sqrt{\rho_1}}\right\}\right]^2,\quad q_0\equiv\frac{|\varepsilon_s|}{\hbar\omega},$$

where $K_{1/3}(x)$ is Basset's function. Taking into account the asymptotic expansion[18] $K_{1/3}(x) \approx (\pi/2x)^{1/2}e^{-x}$, we obtain

$$J_{n_0}^2(\rho_1)=\frac{\hbar\omega^2}{\pi e_0F}\sqrt{\frac{2m}{|\varepsilon_s|}}\exp\left\{-\frac{4}{3}\frac{\sqrt{2m}|\varepsilon_s|^{3/2}}{e_0\hbar F}\right\}. \quad (41)$$

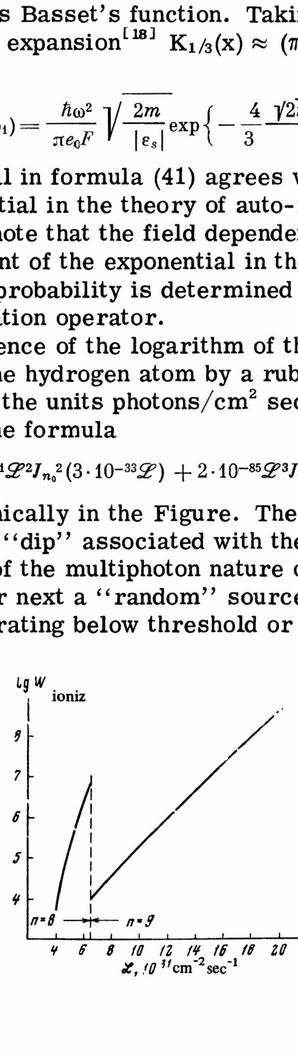
The exponential in formula (41) agrees with the well-known exponential in the theory of auto-ionization of an atom.[19] We note that the field dependence of the factor standing in front of the exponential in the expression for the ionization probability is determined by the choice of the perturbation operator.

The dependence of the logarithm of the probability of ionization of the hydrogen atom by a ruby laser on the intensity $\mathscr{L}$ (in the units photons/cm² sec), calculated according to the formula

$$W_{\text{ioniz}}\approx10^{-51}\mathscr{L}^2J_{n_0}^2(3\cdot10^{-33}\mathscr{L})+2\cdot10^{-85}\mathscr{L}^3J_{m_0}^2(3\cdot10^{-33}\mathscr{L}).$$

is shown graphically in the Figure. The graph has a characteristic "dip" associated with the transition to a higher-order of the multiphoton nature of the process.

We consider next a "random" source (G). This may be a laser operating below threshold or a thermal source



from which a spectrally narrow collimated beam is selected with the aid of a linear filter. (The output distribution of the filter, of course, should possess the specified line contour.) Such sources are essentially narrow band quantum-mechanical generators of noise. In this connection for light the conditions for first-order coherence are assumed to be satisfied (factorization of the correlation functions of first order in the field operators), but the conditions for coherence of higher orders (factorization of the higher-order correlation functions)[11] is not satisfied.

The weight function $\mathscr{P}(\{r_\kappa\})$ for the radiation being investigated may be chosen in the form of a product of Gaussian functions of the individual modes

$$\mathscr{P}^{(G)}(r_\varkappa) = \frac{1}{\pi \bar{n}_{\varkappa_0\lambda_0}} \exp\left\{-\frac{r_\varkappa^2}{\bar{n}_{\varkappa_0\lambda_0}}\right\}. \quad (42)$$

Calculation of $\varphi_{l_1 l_2}$ in Eq. (22) with the function (42) gives

$$\varphi_{l_1 l_2}^{(G)} = (2l_2 + l_1 + \nu)!\,(\bar{n}_{\varkappa_0\lambda_0})^{2l_2+l_1+\nu}, \quad \nu = 2,3. \quad (43)$$

Let us represent the factor $(2l_2 + l_1 + \nu)!$ appearing in Eq. (43) in the form of an integral:

$$(2l_2 + l_1 + \nu)! = \int_0^\infty e^{-\tau}\tau^{2l_2+l_1+\nu}\,d\tau.$$

In addition, for convenience let us write $W^{0(L)}_{\text{ioniz}} = W^{L(2)}_{\text{ioniz}} + W^{L(3)}_{\text{ioniz}}$ from formula (36) as a function of the parameters ρ_1 and ζ:

$$\hbar\omega q = \varepsilon_s + \zeta, \; \zeta = e_0^2 F^2/4m\omega^2. \quad (44)$$

It is not difficult to verify that

$$W^{0(G)}_{\text{ioniz}} = \int_0^\infty e^{-\tau}\tau^2 W^{L(2)}_{\text{ioniz}}(\rho_1\tau;\zeta\tau)\,d\tau + \int_0^\infty e^{-\tau}\tau^3 W^{L(3)}_{\text{ioniz}}(\rho_1\tau;\zeta\tau)\,d\tau. \quad (45)$$

One can only accomplish the integration over τ in the general case by numerical methods. In order to obtain a visible result, we make the following two approximations. Side by side with the ionization potential we neglect the average oscillation energy of the electron in the field of the electromagnetic wave, i.e., we omit $e_0^2 F^2/4m\omega^2$ together with the quantity $|\epsilon_s|$ in formula (44). (Correspondingly we neglect $\zeta\tau$ in Eq. (45) since values of $\tau \lesssim 1$ are essential.) In addition, we confine ourselves to the case $\rho_1 \ll 1$ when, according to the multiplication theorem for cylindrical functions[18]

$$J_{n_0}(\rho_1\tau) \approx \tau^{n_0} J_{n_0}(\rho_1).$$

To the approximations being used, we obtain

$$W^{0(G)}_{\text{ioniz}} \approx n!\,W^{0(L)}_{\text{ioniz}}. \quad (46)$$

From formula (46) it follows that the probability of ionization of an atom per unit time associated with steady-state irradiation by a "random" source is n! (n denotes the number of photons absorbed) times larger than that associated with radiation by an artificial emitter. For the case n = 2, this result is well known.[20,21]

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