

CONTRIBUTION OF COLLISIONS TO MULTIPHOTON RESONANCES IN GASES

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The collision between a slow electron or neutral atom and atoms of a gas in the field of a strong electromagnetic wave is considered. The cross section for the impact-multiphoton excitation mechanism for an hydrogen atom colliding with an electron is calculated. The energy distribution of scattered electrons as a function of electromagnetic radiation frequency and intensity is obtained. The collision cross section and mean excitation probability per unit time are calculated for the case of collisions between neutral atoms in a Maxwellian gas. It is shown that with increasing gas pressure the impact-multiphoton ionization mechanism predominates over the direct multiphoton ionization mechanism.

1. To observe multiphoton resonances in rarefied gases, it is usually necessary to make special changes in the frequency of the laser radiation. On the other hand, as noted by one of the authors^[1] (see also^[2]), multiphoton resonance can be produced in a real gas by compensating the detuning from resonance by the kinetic energy of the gas particles colliding with the absorbing atoms. Inelastic collision of an electron with an atom, accompanied by absorption of several photons, is analogous in many respects to the excitation of an impurity atom in a crystal by electron impact, with simultaneous absorption of many phonons^[3]. The second-order process with absorption of one photon upon collision of an electron with an atom was investigated by Buĭmistrov^[4] within the frame of perturbation theory.

The purpose of the present note is to derive formulas for the probability of multiphoton absorption of light by an atom, with allowance for its collisions with the particles in the gas. It is assumed that the concentration of the gas particles is such that the average time τ between two collisions is shorter than the duration τ_{imp} of the laser pulse yet exceeds the period $2\pi\omega$ of the electromagnetic radiation, i.e.,

$$2\pi/\omega < \tau < \tau_{imp}.$$

It is also assumed that the width of the resonant level of the excited atom, including that part of the width which is due to collisions, is much less than the deviation from resonance.

2. We consider first an inelastic collision of an electron with a hydrogen-like atom interacting with a strong electromagnetic radiation. Let the energy $\hbar\omega$ of m photons be close to the energy difference between the ground state g (with energy ϵ_g) and of the excited state f (with energy ϵ_f) of the atom, and let $\Delta_m = \epsilon_f - \epsilon_g - m\hbar\omega$ be the corresponding deviation from resonance, with $|\Delta_m| > \gamma_f$, where γ_f is the width of the level f . It is obvious that electrons with kinetic energy $\epsilon_{k_0} > \Delta_m$ are capable of compensating for the deviation from resonance and realizing an impact-multiphoton excitation mechanism. The cross section of such an elementary process can be calculated in the Born approximation, a criterion for which, in the case of in-

elastic scattering of an electron with change of energy $\Delta\epsilon$, is^[5,6]

$$\epsilon_{k_0} \gg \Delta\epsilon. \tag{1}$$

In the investigated case we have $\Delta\epsilon = |\Delta_m|$. Having in mind subsequently excitation of optically allowed states of the atom, we neglect for simplicity the exchange interaction, which is not very important in this case^[7].

The probability amplitude $A_{gk_0}^{f\alpha k}(t)$ of the process that causes the atom to go from the ground (g) to the excited (f) state and the electron to change its momentum ($k_0 \rightarrow k$) is determined in the Born approximation by the formula

$$A_{gk_0}^{f\alpha k}(t) = \frac{1}{i\hbar} \int_{-\infty}^t dt' \int d\mathbf{r}_a \int d\mathbf{r} \varphi_k^*(\mathbf{r}, t') \Psi_{f\alpha}^*(\mathbf{r}_a, t') V(\mathbf{r}_a, \mathbf{r}) \Psi_g(\mathbf{r}_a, t') \varphi_{k_0}(\mathbf{r}, t');$$

$$V(\mathbf{r}_a, \mathbf{r}) = \frac{e_0^2}{|\mathbf{r} - \mathbf{r}_a|}. \tag{2}$$

$\Psi_g(\mathbf{r}_a, t)$ is the wave function of the ground state of the atom, which we assume for simplicity to be unperturbed by the electromagnetic field $\Psi_g(\mathbf{r}_a, t) = \psi_g(\mathbf{r}_a) \exp(-i\epsilon_g t/\hbar)$, $\Psi_{f\alpha}(\mathbf{r}_a, t)$ is the wave function of the degenerate level with principal quantum number n_f of a hydrogen atom in an electromagnetic field (α numbers the degenerate states)^[8,9]:

$$\Psi_{f\alpha}(\mathbf{r}_a, t) = \psi_{f\alpha}(\mathbf{r}_a) \exp\{-i\epsilon_f t/\hbar - i\rho_{f\alpha} \sin \omega t\}, \tag{3}$$

where $\psi_{f\alpha}(\mathbf{r}_a)$ are linear combinations of the components of the degenerate level f , and diagonalize the matrix of the dipole ($\sim \mathbf{r}_a$) perturbation. For a z -polarized electromagnetic wave, $\psi_{f\alpha}(\mathbf{r}_a)$ coincides with the wave function of the hydrogen atom in parabolic coordinates

$$\rho_{f\alpha} = \frac{3}{2} \frac{e_0 F a_B}{\hbar \omega} n_1 (n_1 - n_2),$$

$n_1 + n_2 = n_f - 1$, F is the field intensity of the electromagnetic wave, a_B is the Bohr radius, $\varphi_k(\mathbf{r}, t)$ is the wave function of the free electron in the electromagnetic field (see, for example,^[11]):

$$\varphi_k(\mathbf{r}, t) = N_k e^{-i\mathbf{k}\cdot\mathbf{r}} \exp\left\{-\frac{i}{\hbar} (\epsilon_k + \epsilon_p) t + i \frac{e_p}{2\hbar\omega} \sin 2\omega t - i\rho(k) \cos \omega t\right\}, \tag{4}$$

$\epsilon_F = e_0^2 F^2 / 4\mu\omega^2$ is the average kinetic energy of the electron in the field of the electromagnetic wave, $\rho(\mathbf{k}) = e_0 \mathbf{F} \mathbf{k} / \mu\omega^2$, $\epsilon_k = \hbar^2 k^2 / 2\mu$, N_k is a normalization constant, and e_0 and μ are the charge and mass of the electron.

Omitting the intermediate steps, we write down the formula for the cross section of the process in question

$$d\sigma_{gf}(k_0) = \sum_{m=m_0}^{\infty} d\sigma_{gf}^{(m)}(k_0); \quad (5)$$

$$d\sigma_{gf}^{(m)}(k_0) = 4 \left(\frac{\mu e_0^2}{\hbar^2} \right)^2 \sum_{\alpha} J_m^2(\rho_{f\alpha m}) |V_{f\alpha, g}(\boldsymbol{\kappa}_m)|^2 \frac{k_0}{k_m \kappa_m^4} d\alpha; \quad (6)$$

$$V_{f\alpha, g}(\boldsymbol{\kappa}_m) = \int d\mathbf{r}_a \exp(-i\boldsymbol{\kappa}_m \mathbf{r}_a) \psi_{f\alpha}^*(\mathbf{r}_a) \psi_g(\mathbf{r}_a),$$

$$\rho_{f\alpha m} = [\rho_{f\alpha}^2 + \rho^2(\boldsymbol{\kappa}_m)]^{1/2};$$

$$\boldsymbol{\kappa}_m = \mathbf{k}_0 - \mathbf{k}_m, \quad k_m = [k_0^2 - 2\mu\hbar^{-2}(\epsilon_f - \epsilon_g - m\hbar\omega)]^{1/2};$$

$$m_0 = \langle (\hbar\omega)^{-1} [\hbar^2 k_0^2 / 2\mu - (\epsilon_f - \epsilon_g)] \rangle.$$

here $\langle A \rangle$ is the integer part of the number A , $J_\nu(\rho)$ is a Bessel function of real argument, $d\alpha$ is the solid-angle element for the scattered electron. Thus, the cross section $d\sigma_{gf}(k_0)$ for the excitation of an atom in inelastic scattering of an electron in the field of intense electromagnetic radiation is a sum of partial cross sections $d\sigma_{gf}^{(m)}(k_0)$ corresponding to processes in which m photons are absorbed or emitted in the scattering. Each of these processes has its own excitation threshold at an incident particle energy

$$\epsilon_{\text{thr}}^{(m)} = \epsilon_f - \epsilon_g + m\hbar\omega, \quad m = 0, \pm 1, \pm 2, \dots$$

Accordingly, the particle acquires as a result of the scattering an energy

$$\epsilon_k^{(m)} = \epsilon_{k_0} - (\epsilon_f - \epsilon_g) + m\hbar\omega, \quad m = 0, \pm 1, \pm 2, \dots$$

when $\rho_{f\alpha m} \ll 1$, $d\sigma_{gf}^{(m)} \sim \mathcal{L}^{|m|}$ (\mathcal{L} is the density of the electromagnetic radiation flux ($\mathcal{L} \sim F_0^2$)) and the contribution of the m -th partial cross section decreases sharply with increasing $|m|$. However, the condition $\rho_{f\alpha m} \gtrsim 1$ can be realized with increasing field intensity F , and the ratio of the partial cross sections is significantly altered. Thus, a much smaller contribution is made by the partial cross section $d\sigma_{gf}^{(m=0)}$

corresponding to the scattered-electron energy

$\epsilon_k^{(m=0)} = \epsilon_{k_0} - (\epsilon_f - \epsilon_g)$ and hence corresponding to the threshold $\epsilon_{\text{thr}}^{(m=0)} = \epsilon_f - \epsilon_g$, which obtain also in the absence of electromagnetic radiation, so that

$d\sigma_{gf}^{(m=0)} \sim J_0^2(\rho) \ll 1$, $\rho > 1$ (an effect analogous to the

“suppression” of the light absorption at the electron-transition frequency by intense external radiation^[12]).

The parameter $\rho_{f\alpha m}$ increases with increasing scattering angle ϑ (via $\rho(\boldsymbol{\kappa}_m)$, $\kappa_m^2 = k_0^2 + k_m^2 - 2k_0 k_m \cos \vartheta$). At certain scattering angles ϑ , the Bessel function $J_m(\rho_{f\alpha m})$ can go through zero, and this causes vanishing of the corresponding partial cross section $d\sigma_{gf}^{(m)}(k_0)$. Since the Bessel function takes on zero value for different m at different ϑ , electrons with different energies $\epsilon_k^{(m)}$ are scattered at different angles.

At small deviations from resonance Δm ($\Delta m \ll \hbar\omega \ll \epsilon_f - \epsilon_g$) the condition (1) can be satisfied at

rather small incident-electron energy, including also $\epsilon_{k_0} < \epsilon_f - \epsilon_g$. In this interesting case, formula (6) describes the process of multiphoton excitation of the hydrogen atom, when the deviation from resonance is compensated for by the kinetic energy of a slow (say, thermal) electron ($\rho_{f\alpha m} \approx |\rho_{f\alpha}|$). This process is then the only possible mechanism for exciting the atom. Neither radiation (since resonance conditions are not satisfied) nor a thermal electron (because of the low kinetic energy) is capable of exciting the atom separately.

The process considered above may turn out to be a realistic mechanism for laser-radiation absorption in a weakly ionized gas under conditions preceding laser breakdown (i.e., at not too high radiation intensities and laser-pulse durations). In this case the known competing mechanism of the inverse bremsstrahlung effect is not significant^[13].

3. We consider now the impact-multiphoton mechanism of excitation in a gas with neutral atoms. In this case, the deviation from resonance is compensated for by the kinetic energy of the colliding atoms. A detailed quasiclassical theory of inelastic atomic collisions has already been developed (see^[14, 15]). In many cases in the calculation of the cross sections of the inelastic processes in the collisions of heavy particles one uses the so-called linear-term model, which leads in final analysis to the Landau-Zener formula. We shall calculate later on the cross section of the process of interest to us within the framework of this model.

We consider the collision of two generally speaking different atoms A and B, as a result of which one of the atoms (for example, A) goes over to an excited state ($\epsilon_f^{(A)}$) because of absorption of m photons of radiation and because of the decrease of the kinetic energy of the relative motion of the partners A and B. Kinetic energy must be consumed to compensate for the deviation from resonance $\Delta_m = \epsilon_f^{(A)} - \epsilon_g^{(A)} - m\hbar\omega$.

We confine ourselves to the case of a doubly degenerate state f of a hydrogen-like atom (for example, the level with $n_f = 2$ of the hydrogen atom with components $2s = 1$, $2p = 2$). The corresponding wave functions $\Psi_{f(\pm)}(\mathbf{r}_A, t)$, according to (3), are given by

$$\Psi_{f(\pm)}(\mathbf{r}_A, t) = 2^{-1/2} (\psi_1(\mathbf{r}_A) \pm \psi_2(\mathbf{r}_A)) \exp\{-i\epsilon_f t / \hbar \pm i\rho_f \sin \omega t\},$$

$$\rho_f = 3e_0 F a_B / \hbar\omega.$$

We seek the wave function of the system of colliding partners in the form

$$\Phi(\mathbf{r}_A, \mathbf{r}_B, R, t) = a_0(R) \Psi_g(\mathbf{r}_A, t) \Psi_g(\mathbf{r}_B, t) +$$

$$+ a_{(+)}(R) \Psi_{f(+)}(\mathbf{r}_A, t) \Psi_g(\mathbf{r}_B, t) +$$

$$+ a_{(-)}(R) \Psi_{f(-)}(\mathbf{r}_A, t) \Psi_g(\mathbf{r}_B, t).$$

The subscript of the coordinate \mathbf{r} identifies the atom to which it belongs, while $R = R(t)$ is the distance between the centers of the partners, which is a classical trajectory of their relative motion.

We expand $\Psi_{f(\pm)}(\mathbf{r}_A, t)$ in a Fourier series and confine ourselves to terms corresponding to the minimum deviation Δ_m from m -photon resonance. We introduce the notation

$$\bar{a}_0(R) = a_0(R) \exp\left\{-\frac{i}{\hbar} \int v_{gg}(R) dt\right\},$$

$$\tilde{a}_{(\pm)}(R) = a_{(\pm)}(R) \exp \left\{ -\frac{i}{2\hbar} \int [v_{11}(R) + v_{22}(R) \pm v_{12}(R)] dt \right\};$$

$$v_{ij}(R) = \int \Psi_i^*(\mathbf{r}_A) \Psi_j^*(\mathbf{r}_B) H_{AB}(\mathbf{r}_A, \mathbf{r}_B, R) \Psi_j(\mathbf{r}_B) \Psi_i(\mathbf{r}_A) d\mathbf{r}_A d\mathbf{r}_B,$$

where $H_{AB}(\mathbf{r}_A, \mathbf{r}_B, R)$ is the energy of interaction of the atoms A and B, which are separated by a distance R. The coefficients $\tilde{a}_0(R)$ and $\tilde{a}_{\pm}(R)$ satisfy the system of equations

$$\begin{aligned} i\hbar \dot{\tilde{a}}_0(R) &= \frac{1}{2^{1/2}} v_{g2}(R) J_{-m}(\rho_f) \exp \left\{ -\frac{i}{\hbar} \int (\Delta_m + \Delta U_{(+)}(R)) dt \right\} \tilde{a}_{(+)}(R) \\ &+ \frac{1}{2^{1/2}} v_{g2}(R) J_m(\rho_f) \exp \left\{ -\frac{i}{\hbar} \int (\Delta_m + \Delta U_{(-)}(R)) dt \right\} \tilde{a}_{(-)}(R), \quad (7) \\ i\hbar \dot{\tilde{a}}_{(+)}(R) &= \frac{1}{2^{1/2}} v_{2g}(R) J_{-m}(\rho_f) \exp \left\{ \frac{i}{\hbar} \int (\Delta_m + \Delta U_{(+)}(R)) dt \right\} \tilde{a}_0(R); \\ i\hbar \dot{\tilde{a}}_{(-)}(R) &= \frac{1}{2^{1/2}} v_{2g}(R) J_m(\rho_f) \exp \left\{ \frac{i}{\hbar} \int (\Delta_m + \Delta U_{(-)}(R)) dt \right\} \tilde{a}_0(R); \\ \Delta U_{(\pm)}(R) &= v_{11}(R) + v_{22}(R) \pm v_{12}(R) - v_{g2}(R). \end{aligned}$$

In formulas (7) we have omitted, for simplicity, the matrix element $v_{g1}(R)$, which connects states of equal parity of the atom A, in comparison with $v_{g2}(R)$, which connects states of different parity. For the same reason, $v_{12}(R) > v_{gg}(R)$, $v_{11}(R)$, $v_{22}(R)$. Consequently, $v_{12}(R)$ is the principal term determining the vanishing of the expressions $\Delta_m + \Delta U_{(\pm)}(R)$ ("crossing" of the adiabatic potentials). Obviously, the "crossing" is possible only for $\Delta U_{(-)}(R)$ in the case when the Massey parameter is $\Delta m a_B / \hbar v \gg 1$ (v is the velocity of the relative motion of the colliding atoms), the corresponding terms in (7) make the principal contribution. In this case ($\Delta m a_B / \hbar v \gg 1$) one should also expect the linear-term operator to give a satisfactory result. We expand $\Delta_m + \Delta U_{(-)}(R)$ near the "crossing" point R_C , determined from the condition $\Delta_m + \Delta U_{(-)}(R_C) = 0$:

$$\begin{aligned} \Delta_m + \Delta U_{(-)}(R) &\approx \Delta Q(R_C) (R - R_C), \\ \Delta Q(R_C) &= \frac{\partial}{\partial R} \Delta U_{(-)}(R) |_{R=R_C}. \end{aligned}$$

The system (7), under the simplifying assumptions made above, takes the form

$$\begin{aligned} i\hbar v_R \frac{d\tilde{a}_0}{dx} &= \frac{1}{2^{1/2}} v_{g2}(R_C) J_m(\rho_f) \exp \left\{ i \frac{\Delta Q(R_C)}{2\hbar v_R} x^2 \right\} \tilde{a}_{(-)}, \quad (8) \\ i\hbar v_R \frac{d\tilde{a}_{(-)}}{dx} &= \frac{1}{2^{1/2}} v_{g2}(R_C) J_m(\rho_f) \exp \left\{ -i \frac{\Delta Q(R_C)}{2\hbar v_R} x^2 \right\} \tilde{a}_0. \end{aligned}$$

Here $x = R - R_C$, and v_R is the value of the radial component of the velocity of the relative motion at the "crossing" point R_C ($v_R = \partial R / \partial t |_{R=R_C}$).

A solution of the system (8) is known (see, for example^[15], p. 113). The probability \mathcal{P} of the process of interest to us, obtained with the aid of the solution of (8), is expressed by the Landau-Zener formula:

$$\mathcal{P} = 2e^{-1} (1 - e^{-1}).$$

The parameter ξ takes in this case the form

$$\xi = \frac{\pi v_{g2}^2(R_C)}{\Delta Q(R_C) \hbar v_R} J_m^2(\rho_f) = \xi_0 J_m^2(\rho_f).$$

The cross section of the process is determined by the formula

$$\sigma_m = 2\pi \int_0^{b_{\max}} \mathcal{P}(b) b db, \quad (9)$$

b is the impact parameter. \mathcal{P} depends on the impact

parameter via the radial velocity

$$v_R = \left(\frac{2}{M} \right)^{1/2} \left[(\epsilon - U_C) - \frac{b^2}{R_C^2} \epsilon \right]^{1/2}$$

(U_C is the potential energy of the interaction of the colliding atoms at the "crossing" point, $\epsilon = Mv^2/2$, M is the reduced mass of the partners). The integration in (9) is carried out up to an impact-parameter value b_{\max} , at which v_R vanishes (see^[10], p. 389). If $\rho_f < m$, $J_m^2(\rho_f) \ll 1$ we have in this case $\xi < 1$ even if $\xi \gtrsim 1$. Then

$$\mathcal{P} = \frac{2\pi v_{g2}^2(R_C)}{\Delta Q(R_C) \hbar v_R} J_m^2(\rho_f). \quad (10)$$

We note that in this case formula (10) can be obtained by approximately solving the system (m), if we put $\tilde{a}_0(R) \sim 1$. In the same approximation $J_m^2(\rho_f) \ll 1$, $\tilde{a}_0(R) \sim 1$ we can consider the more complicated case of excitation of an atomic level of arbitrary degeneracy multiplicity, and also a case frequently encountered in real atoms, that of quasidegenerate levels that are strongly mixed by electromagnetic radiation, when the deviation from resonance with any of these levels is smaller or larger than the energy distance between levels. In all these cases, a formula of the type (10) is obtained for the probability of the impact-multiphoton excitation.

Substitution of (10) in (9) yields

$$\sigma_m(\epsilon) = \frac{2(2M)^{1/2} \pi^2 v_{g2}^2(R_C) R_C^2 (\epsilon - U_C)^{1/2}}{\hbar \Delta Q(R_C) \epsilon} J_m^2(\rho_f). \quad (11)$$

Of practical interest is knowledge of the average probability of excitation of an atom per unit time W :

$$W = n_0 \int_{v_0}^{\infty} \sigma_m(\epsilon) v f(\epsilon) d\epsilon,$$

where n_0 is the density of the gas particles, $f(\epsilon)$ is their energy distribution. Assuming that the collisions occur in a Maxwellian gas, we find

$$W = \frac{2^{1/2} \pi^{3/2} v_{g2}^2(R_C) R_C^2 n_0}{\hbar \Delta Q(R_C)} J_m^2(\rho_f) \exp(-U_C/kT), \quad (12)$$

k_1 is the Boltzmann constant and T is the absolute temperature. As follows from (12), the probability of impact-multiphoton excitation increases linearly with gas pressure.

For collisions between like atoms, it would be necessary to take formally into account the additional symmetry of the wave function of the system, a symmetry due to the identity of the partners. In the approximations under which formula (10) is obtained, this refinement can be easily introduced and the result is again expressed by a formula of the type (10), consisting of contributions coming from the symmetrical and antisymmetrical parts of the wave function of the system. The situation $\Delta_m < 0$, when the resonance is due to an increase in the kinetic energy of the relative motion of the colliding atoms, can be considered in analogy with the procedure used above.

Let us consider by way of example the excitation of the level $n = 5$ of the hydrogen atom by radiation from a neodymium laser, $\hbar\omega = 1.18$ eV. The states 5s, 5p, 5d, 5f, and 5g are strongly mixed by the electromagnetic radiation. At $F = 10^7$ V/cm we have

$$\max \rho_i \approx 1.35, J_{i1}^2(1.35) \ll 1$$

and we consequently use formula (12), to estimate the excitation probability per unit time. We estimate the parameters in (12) in the following manner^[15]:

$$\Delta Q(R_c) \sim \Delta_m / a_B, v_{\text{eff}}(R_c) \sim \Delta_m / n^{1/2}, \\ U_c \sim \Delta_m, R_c \sim (I_H / \Delta_m)^{1/2} a_B, I_H = e_c^2 / 2a_B.$$

At $n_0 \sim 10^{20} \text{ cm}^{-3}$ and $T = 300^\circ \text{K}$ we obtain $W \sim 10^{-12} \text{ sec}^{-1}$. The probability of ionization of a hydrogen atom per unit time under this condition is also determined by the value of W , since ionization of the resonant level $n = 5$ in a field of such high intensity has a probability close to unity. The probability per unit time of direct multiphoton ionization W_d , estimated from perturbation-theory formulas^[16], yields $W/W_d \sim 10^4$, i.e., the impact-multiphoton mechanism of excitation prevails over the mechanism of direct multiphoton ionization.

It can thus be assumed that the impact-multiphoton mechanism of excitation is responsible for the appearance of free electrons when laser radiation is focused in a pure gas under conditions preceding laser breakdown^[17]. This mechanism can also provide the necessary number of initial electrons to start an electron avalanche in a laser breakdown.

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