Multiphoton Photoeffect and the Theory of the Delay Time

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The reaction rate of the multiphoton photoeffect in an isolated atom is calculated by quantum electrodynamics methods with allowance for the possibility of re-emission of an arbitrary number of photons. If the photon flux $j<j_0=\max\{1/\lambda^2\tau\}$ (τ is the delay time in elastic $e\gamma$-scattering), then a tunnel multiphoton photoeffect occurs, and if $j>j_0$, a suprabarrier multiphoton photoeffect occurs without re-emission (saturation). It is shown that previous works yielded low values of the reaction rates, since classical values of τ, or values in the threshold value of j_0 which were not consistent with the structure of the atom, appeared in the calculations.

1. INTRODUCTION

THE probability of the multiphoton photoeffect (MPPE) on an isolated atom has been calculated by different methods and by many workers[1-7]. The results of these calculations, however, are contradictory and do not agree, generally speaking, with experiment (for example,[8]). In the present article[1)] the MPPE reaction rate was calculated by a method developed by one of the authors[10], with an arbitrary number of reradiated photons taken into account in the reaction rate (Sec. 2) or in the S matrix (Sec. 3). This leads to expressions that are close in form, in corresponding cases, to the results of[1-3], but differ greatly from them numerically. It is shown in Sec. 4 that in some of the cited papers[1-3,7] the formulas have a semiclassical character, since they correspond to the assumption that the $e\gamma$ interaction occurs only in classical regions; the formulas of other papers[4-6] correspond to quantum broadening of the interaction region, but without allowance for the possibility of reradiation of the photons.

The analysis of MPPE (as well as of other multiphoton processes) is complicated by the fact that when the flux density j increases, certain types or stages of the interaction become saturated (the transition probability reaches unity), and new reaction channels are opened[10]. Thus, for example, for direct absorption of two photons it is necessary to satisfy the relation

$$j > j_0 \approx 1/\lambda^2\tau, \tag{1.1}$$

where τ is the delay time in the scattering of the first photon by an electron. If $j < j_0$, then it is necessary to take into account in the absorption of two photons the possibility of absorption and reradiation of additional quanta. Thus, if $j > j_{0\,max} = 1/\lambda^2\tau_{min}$ (τ_{min} is the smallest time delay in the intermediate stages of the MPPE), then the MPPE proceeds as a direct (suprabarrier) process with unity probability, and there is no need to take into account the possibility of reradiation in it. On the other hand, if j is smaller than some threshold value j_{0k}, then the MPPE is a tunnel transition, calculated with allowance for reradiations in this stage. If $j > j_{0k}$ ($k \leq n$) and $j < j_l$ ($l > n$), then we can assume that the electron is at all times in the state n, and the MPPE must be calculated from the corresponding level.

The delay time in scattering with angular momentum l is determined in terms of the partial S matrix

$$\tau_l = \mathrm{Re}\,\frac{d\ln S_l}{id\omega}. \tag{1.2}$$

For potential scattering, when the S matrix can be represented, according to Hoo and Regge (see, for example,[11]) in the form of Blaschke products with stable particles and resonances (realizable in the intermediate state), and also in our case, expression (1.2) can be approximately written in the form

$$\tau_l = \frac{1}{\omega} + \sum_n \frac{\Gamma_n(l)/2}{(\omega-\omega_n(l))^2 + \Gamma_n^2(l)/4}. \tag{1.3}$$

Here $\Gamma_n(l)$ and $\omega_n(l)$ are the widths and frequencies of the levels with specified l, natural in the case of a weak field or broadened and shifted in a strong field. (Although Γ_n and ω_n can be calculated in the presence of a field, in the present paper we assume for them the experimental values.) For a free electron, according to (1.3), in accordance with the uncertainty principle, $\tau \approx 1/\omega$ and at $j > c\lambda^{-3}$ it is necessary to take into account in the calculation of the MPPE the acceleration of the electron in the field of the wave. Choosing $j < c\lambda^{-3}$ (we confine ourselves just to this case) we can separate the MPPE from processes with the free electron[2)].

2. CALCULATION OF REACTION RATE

Let us calculate the reaction rate of an N-quantum photoeffect on an isolated atom with allowance for reradiation ($n \gg N$):

$$(n+N)\gamma(\omega) + e_0 \to n\gamma(\omega) + e_f. \tag{2.1}$$

The S matrix of the reaction (2.1) is considered in the lowest, (2n + N)-th order, and the potential A_μ is expanded in plane waves in the volume V. Allowance for the monochromaticity of the incident radiation leads for each absorbed and emitted photon, respectively, to the substitutions

$$\frac{dn_i}{V} \to f(\omega, \mathbf{k}_i)\,d\mathbf{k}_i \to \frac{j}{c}\,\delta(\mathbf{k}_i - \mathbf{k}_0)\,d\mathbf{k}_i, \quad \frac{1}{V} \to \frac{d\mathbf{k}_f}{(2\pi\hbar)^3}. \tag{2.2}$$

[1)]Some preliminary results were already reported earlier[9].

[2)]Relations of the type (1.1) result from the standard calculation of the rate of the transition[10]. The concept of delay time is in this case the only possible physical interpretation of the singularities of the reaction rates.

Considering only dipole transitions, we can express all the matrix elements (except for the last one, corresponding to a transition from the virtual state into the continuous spectrum with absorption of one quantum) in terms of the total level widths and the transition frequencies:

$$\int d\Omega |\langle n|\gamma_k e^{i\mathbf{kr}}|m\rangle|^2 \to \frac{8\pi^2\Gamma_n}{e^2\omega_{nm}}. \quad (2.3)$$

In view of the large level density at the boundaries of the continuous spectrum, the absorption of the last of the N quanta occurs near some real level, and the corresponding matrix element M_f can be expressed approximately in terms of the known oscillator strengths. Thus, for two-quantum ionization of the hydrogen atom from the 2s state by ruby-laser light, this transition can be regarded as the electronic transition 3p → ∞d, and we can accordingly assume[12]

$$\Sigma |M_f|^2 \to \frac{2}{3}\cdot 5{,}46\left(\frac{\hbar}{me^2}\right)^2. \quad (2.4)$$

(The fact that the electron in the intermediate state is not exactly at the 3p level is taken into account by the δ function of the energy conservation law in the S matrix). We shall carry through the calculation for this case of ionization to conclusion. Taking into account only the diagrams closest to the resonance, we obtain for the rate of the reaction (2.1) in accordance with[10], the expression

$$R_n = \frac{2\pi}{e^2} W \left(\frac{j}{4j_0}\right)^{N-2} (n+1)\left(\frac{\pi}{2}\frac{j}{j_0}\right)^{n+1}, \quad (2.5)$$

where we have omitted the factors corresponding to the multipolarity of the electron, and all the factors corresponding to the last transition are concentrated in W. The factor j_0, which plays the role of the critical flux density, is obtained by substituting (1.3) in (1.1):

$$j_0 = 1/\lambda^2\tau \sim 2[(\omega-\omega_{3p})^2 + \Gamma_{3p}^2/4]/\lambda^2\Gamma_{3p}. \quad (2.6)$$

The probability W is defined as

$$W = j\frac{e^2}{(2\pi)^2\omega}\sum_{n_i, n_f} |M_f|^2\delta(E_{inter} + \omega - E_f)\,d\mathbf{p}$$
$$\to j(e^2/2\pi)\omega m[2m(I - N\hbar\omega)]^{1/2}|\mathbf{r}|^2$$
$$= A(\omega/2\pi)(\hbar c/e^2)[2(I - N\hbar\omega)/mc^2]^{1/2}, \quad (2.7)$$

where for the transition 3p → ∞d, with allowance for (2.4), the coefficient is A = 3.64, and E_{inter} is the energy of the intermediate state, which is close to $\hbar\omega_{3p}$.

The total transition rate is obtained for $j < j_0$ by summing (2.5) over n, i.e., by taking into account an arbitrarily large number of beats of the electron between the 2s level and a virtual level close to 3p (tunneling or sub-barrier ionization):

$$R = C\omega[(I - N\hbar\omega)/mc^2]^{1/2}(j/4j_0)^N(1 - \pi j/8j_0)^{-2}, \quad (2.8)$$

where $C = A(\pi 2^{1/2}/64e)(4\pi\hbar c/e^2) \approx 10$. At $j > j_0$, Eq. (2.8) is not correct, since the ionization already proceeds directly from the 3p level.

If N > 2, then the calculation becomes much more complicated, since different numbers of reradiations are possible at different stages of MPPE.

In the general case, the matrix element is a sum of expressions corresponding to all possible variants of the process; the expression for each variant, in turn, is the product of the matrix elements for the individual transitions with reradiations, raised to powers equal to the numbers of such reradiations. If we disregard the interference between the individual transitions (see Sec. 3), then the rate R_n of a reaction of order 2n + N is proportional to the expression

$$j^{n+N}\sum_{k_1,\ldots,k_{N-1}} (j_{01}^{k_1}\ldots j_{0\,N-1}^{k_{N-1}})^{-1}, \quad k_1+\ldots+k_{N-1}=n+N, \quad (2.9)$$

where $j_{0q} = 1/\lambda^2\tau_q$. The influence of all the levels of the atom is taken into account in (2.9) because of the dependence of j_{0q} on τ_l (1.3), in full accord with[6].

3. ALLOWANCE FOR THE INTERFERENCE TERMS

Near the resonance, as shown in[10], the interference between the matrix elements with different numbers of real (re-emitted) photons is small. For the reactions (2.1), however, in which the intermediate states are far from resonance, the interference terms may turn out to be appreciable. In summing the matrix elements (2.1) over n, we assume in analogy with (2.2) the following conditions on the quantization volumes:

$$1/V_f \to (2\pi\hbar)^{-3},\ 1/V_i \to j/c. \quad (3.1)$$

The matrix element (2.1) at N = 2 can be rewritten in the form

$$M_{2n+2} = -i\frac{n!(n+2)!}{(2n+2)!}\langle f|\hat{e}e^{i\mathbf{kR}}|2n+1\rangle\left(\frac{j}{2\omega c}\right)^{1/2} z^{2n+1}, \quad (3.2)$$

$$z^2 = -e\left(\frac{j}{4\omega^2 c}\right)^{1/2}\left[\sum_k \frac{\langle k|\hat{e}_i e^{i\mathbf{kR}}|k+1\rangle}{E_{kk'} - \omega - i\Gamma_k/2}\right]^2. \quad (3.3)$$

Then the total matrix element is

$$M = \sum_{n=0}^{\infty} M_{2n+2} = M_2\sum\frac{n!(n+2)!}{(2n+2)!}z^{2n}$$
$$= M_2\frac{\pi}{8z^2}\frac{d}{dz}\left[z^3\left(1-\frac{z^2}{4}\right)^{-1/2}\arcsin\frac{z}{2}\right]. \quad (3.4)$$

The expression for the reaction rate has poles in z^2:

$$R \approx |M|^2 \sim [(4 - \mathrm{Re}\,z^2)^2 + (\mathrm{Im}\,z^2)^2]^{-1/2}, \quad (3.5)$$

where

$$\mathrm{Re}\,z^2 = \sum D_k\frac{(\omega_{kk'}-\omega)^2 - (\Gamma_k/2)^2}{[(\omega_{kk'}-\omega)^2 - (\Gamma_k/2)^2]^2 + [\Gamma_k(\omega_{kk'}-\omega)]^2},$$
$$\mathrm{Im}\,z^2 = \sum D_k\frac{\Gamma_k(\omega_{kk'}-\omega)}{[(\omega_{kk'}-\omega)^2 - (\Gamma_k/2)^2]^2 + [\Gamma_k(\omega_{kk'}-\omega)]^2},$$
$$D_k \equiv -e(j/4\omega^2 c)^{1/2}|\langle f|\hat{e}e^{i\mathbf{kR}}|2n-1\rangle|^2 \to \pi\Gamma_k(j/\omega^2\omega_{kk'}^2). \quad (3.6)$$

As follows from (3.5), the pole is obtained only at resonance (Im $z^2 \to 0$, Re $z^2 \to D\tau^2$), if Re $z^2 = 4$, corresponding exactly to the pole in (2.8) (much more complicated calculations at N > 2 lead to analogous results). Thus, allowance for the possibility of interference does not violate the qualitative conclusions of Sec. 2.

4. DISCUSSION OF RESULTS

1. Our analysis (see also the earlier papers[9,10]) differs from the other approaches in that a new dimensionless expansion parameter has been introduced. Thus, in the well known review by Eberly[13], the following dimensionless parameters were defined (three classical and two quantum)

$$C_1 = j\lambda_c r_0^2/c,\quad C_2 = j\lambda_c r_0\lambda/c,\quad C_3 = j\lambda_c\lambda^2/c,$$

$$Q_1 = j\lambda^3/c, \qquad Q_2 = j\lambda^2 r_0/c, \tag{4.1}$$

where $\lambda_c = \hbar/mc$ and $r_0 = \alpha\lambda_c = e^2/mc^2$. With the aid of the duration τ of the interaction we can define a number of new parameters. For τ itself we can choose any of the following expressions:

$$\tau_l = \sum_n \frac{\Gamma_n/2}{(\omega_{ln}-\omega)^2+\Gamma_n^2/4},$$

$$\tau = 1/\omega,\ \tau' = \hbar/mc^2,\ \tau'' = \alpha\tau' = e^2/mc^3, \ldots, \tag{4.2}$$

or sums of the times (4.2), just as in (1.3). However, only quantities with τ_l lead to new values of the parameters (for bound electrons, when $\tau_l > \tau$). In our calculations this gives rise in natural fashion to the parameter

$$Q_3 = j\lambda^2\tau_l \equiv j/j_0 \tag{4.3}$$

(far from resonance (according to (1.3), $Q_3 \to Q_1$).

2. The absorption and reradiation of laser photons is usually regarded[13] as an "external" renormalization that leads only to a constant factor in the wave function of the electron. Yet, as shown in[10] (see also (2.8)), this additional factor depends strongly on j near the threshold j_0. This is precisely why it appears that the "number of absorbed quanta" defined in (2.8) in terms of $A_j^{k_0}$ is a "good quantum number" only if all the intermediate saturating densities j_{0k} are small: $j_{0k} \ll j$. On the other hand, if any of the stages of the MPPE is close to resonance and $j \gtrsim j_{0res}$, then the MPPE proceeds in fact from this resonant level. It is precisely this circumstance, i.e., the differences in the values of j_{0k}, which explains the discrepancy in the determination of k_0 and the results of the work by G. and N. Delone[14].

3. The quantities τ and j_0 were not introduced by us artificially, but arose (more accurately, were separated) during the course of a standard quantum-electrodynamic calculation. With the aid of τ, j_0, and other observable quantities it becomes possible to describe the results of the experiments. It is therefore natural to assume that these quantities can be separated in the final formulas of other authors, and that the causes of the discrepancies in the values of R can be established by comparison with the values obtained by us. Thus, in his fundamental paper[1], Keldysh introduces the parameter γ, which can be expressed in our notation and with allowance for (4.1) in the form

$$\gamma = (j/j_0^{(N)})^{-1/2} = (Q_2/N)^{1/2}$$

$$j = eE^2/4\pi\hbar c, \qquad j_0^{(N)} = 2\pi/\lambda^2\tau_N, \qquad \tau_N = \tau''/N = e^2/mc^3N. \tag{4.4}$$

When $\gamma \gg 1$, i.e., at $j \ll j_0^{(N)}$), the corresponding formula of Keldysh can be rewritten in a form close to (2.8) and corresponding to allowance for the reradiations:

$$R = \text{const}\,(j/4j_0^{(N)})^N (1 - j/4j_0^{(N)})^{-1}. \tag{4.5}$$

In the spirit of the theory of the delay time, Eq. (4.4) signifies that all the N photons are absorbed simultaneously in a classical volume $\lambda_2 r_0$. Therefore the quantity $j_0^{(N)}$ is much larger than j_0 from (2.8), leading to a strong underestimate of the values of R[8]. In addition, in this approach no account is actually taken of the structure of the atom, which in our case is reflected by the large number of values of τ given by formula (1.3). In fact, Perelomov, Popov, and Terent'ev[3] use expansions in terms of this parameter, and take the structure of the atom into account only by varying the common factor in (4.5). The constructions in the papers of Nikishov and Ritus are similar[2], but in the first of them the expansion parameter is chosen in the form $C_2 = \alpha j/j_0'$, where $j_0' = c/\lambda_c^2\lambda$. It is important to note that the case $\gamma \ll 1$ in[1-3] (i.e., the case $j \gg j_0$) is not comparable with our results, for when $j \gg c/\lambda^3$ the probability of absorption of many photons by a free (ionized) electron is large[9].

In many papers[4-7], no account is taken of the possible reemission of photons, i.e., only the first term of the series (2.8) (or 2.5) at n = 1) is retained. The result of Bebb and Gold[4] corresponds in this case to the choice $j_0' = 1/\lambda^2\langle\tau\rangle$, where $\langle\tau\rangle$ is the delay time averaged over all the levels. This underestimates the value of R[8], owing to the neglect of certain levels[6]. Morton[5] obtained the parameter (4.4) for an interaction Hamiltonian of the $\mathbf{p}\cdot\mathbf{A}$ type, and the parameter (4.4) with the substitution $j \to j\exp(2/N^2)$ when the interaction was chosen in the form $e\mathbf{r}\cdot\mathbf{E}$. Kovarskiĭ's results[7] on multiphoton excitation correspond to the choice of the expansion parameter C_2 from (4.1).

4. All the foregoing shows that introduction of the concepts and values of the delay time and of the saturation threshold into the theory is not a formal device, but is dictated by the entire logic of scattering theory (in any case for multiphoton or multiparticle processes in general). These concepts, and only these, make it possible to determine the dependence of the transition probability on the flux intensity, to subdivide reactions of the MPPE type into saturated and unsaturated stages, to calculate the thresholds of the suprabarrier transitions, etc.

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