Interference phenomena in photoionization-like processes in a group of coherently populated Rydberg levels

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We investigate the dynamics of coherent population of Rydberg atomic levels by a short laser pulse, and the dynamics of resonant ionization with excitation of many Rydberg levels as intermediate states. We also investigate transitions to the continuum or to states of a discrete spectrum from a state that is a coherent superposition of Rydberg-level wave functions and is induced by a radiation pulse of short duration at a definite delay relative to the instant of production of the initial state. We investigate the dependence of the probabilities of these transitions on the delay time and on the duration of the transition-inducing pulse. A number of interference phenomena that lead to an increase of the transition probabilities and to their unusual dependence on the pulse duration are observed, and are found to be due to the formation of packets of coherently populated Rydberg levels.

1. INTRODUCTION

Action of a short pulse of electromagnetic radiation of suitable frequency on an atom can cause coherent population of highly excited, Rydberg, atomic levels. Coherent population means that the ensuing state is described by a wave function (and not by a diagonal density matrix) that is a superposition (packet) of wave functions φ_n of Rydberg levels E_n. Such a state can result either from a direct one-photon transition or from multiphoton excitation. Coherently populated Rydberg levels can assume the role of intermediate levels in multiphoton excitation of higher multiplicity or in multiphoton ionization. A transition from a group of coherently populated Rydberg levels to the continuum or to other states of a discrete spectrum can be effected either by the same field that produces the initial packet of the Rydberg-level wave functions or by another pulse of electromagnetic radiation of different frequency and duration, possibly turned on at some time t_0 later than the instant of production of the initial packet.

Inasmuch coherent population establishes phase relations between the probability amplitudes of finding an atom in states φ_n, it is obvious that in transitions to other states interference can take place between transitions from different Rydberg levels E_n. Since these phase relations vary with time, the degree of interference of the transitions can also depend on the time. This raises the question of identifying the specific manifestations of the discussed interference phenomena, and their dependences on the duration of the pulse that causes the transition, on its delay time t_0, on their localization in the atomic spectrum of the initial level packet, on whether the transition is to higher or lower stats of the discrete spectrum or to the continuum, etc. The present paper deals with these questions.

We consider in the next section the dynamics of coherent population of Rydberg levels acted upon by a short laser pulse, and two-step (resonant) ionization in which the Rydberg levels assume the role of intermediate states. We consider next the photoionization of coherently populated Rydberg levels by another radiation pulse as a function of its duration and delay time, transitions to the continuum, induced recombination, and transitions between groups of Rydberg levels.

2. COHERENT POPULATION OF RYDBERG LEVELS AND TWO-PHOTON IONIZATION

Let an atom in the ground (φ_0) state (with energy E_0) be acted upon by a short radiation pulse of frequency ω and with maximum amplitude F_0 of the electric field strength and with maximum amplitude of the temporal envelope

$$f(t)=\exp(-t^2/\tau^2), \quad \tau=\tau_0/(2\ln 2)^{1/2}, \tag{1}$$

where τ_0 is the pulse duration at half maximum (we shall hereafter designate the pulse duration by τ, bearing in mind the difference between τ and τ_0).

The Gaussian form (1) of the envelope $f(t)$ is the most suitable for single-frequency laser pulses. In the case of atom interaction with a microwave field, other regimes can be realized, in which $f(t)$ is closer to a step function. All the results and qualitative relations obtained below are independent of the form of the envelope. We therefore present below, where possible, general equations with an arbitrary function $f(t)$ as well as (for clarity and to obtain explicit analytic expressions) the corresponding equations corresponding to the Gaussian envelope $f(t)$ of Eq. (1).

Let the frequency ω be lower than the ionization threshold, $\omega < |E_0|$, but close enough to it to permit even one-photon excitation to raise the atom to a Rydberg state φ_0 with energy $E_n = -1/2n^2$ (in atomic units) and with large values $n \gg 1$ of the principle quantum number. The time-dependent probability amplitudes of excitation of Rydberg levels can be easily obtained in first-order perturbation theory in the interaction of the atom with the field $\mathbf{F}_0$:

$$C_n(t)=-i\frac{V_{n0}}{2}\int_{-\infty}^{t} dt' f(t')\exp\{i(E_n-E_0-\omega)t'\}, \tag{2}$$

where $V_{n0} = -\mathbf{d}_{n0}\cdot\mathbf{F}_0$ and $\mathbf{d}$ is the dipole moment of the atom.

The probability of exciting an individual Rydberg level in the entire time of the pulse (1) is equal to

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$$w_n=|C_n(\infty)|^2=\frac{\pi}{4}|V_{n0}|^2\tau^2\exp\left\{-\frac{1}{2}(E_n-E_0-\omega)^2\tau^2\right\}. \tag{3}$$

The total probability of exciting all the Rydberg levels is equal to the sum of the w_n or Eq. (3). According to Ref. 1, the dependence of the matrix elements V_{n0} with $n\gg 1$ on n is given by

$$V_{n0}=V_{10}/n^{3/2}, \tag{4}$$

where V_{10} is the matrix element of the transition from the ground state to the first excited one.

Let the duration of pulse (1) be such that

$$\tau\ll n_0^3, \tag{5}$$

where $n_0=(2|E_0+\omega|)^{-1/2}$ is the principal quantum number corresponding to the center of the band of excited Rydberg levels. The condition (5) means that the width of this band ($\sim 1/\tau$) is much larger than the distance $\Delta_n\equiv n_0^{-3}$ between the closest Rydberg levels. If $\tau\gg n_0^2$, the width of this band is much less than the distance to the threshold of continuum, but if $\tau\ll n_0^2$ the band of excited levels contains also continuum states. In any case, the condition (5) makes it possible to change over in the calculation of the total excitation probability from summation over n to integration with respect to E_n; this yields

$$w=\sum_n w_n=\frac{\pi}{2}V_{10}^2\int_{-\infty}^{+\infty}dt\,f^2(t)$$
$$=\frac{2^{1/2}\pi^{3/2}}{4}V_{10}^2\tau. \tag{6}$$

The linear $w(\tau)$ dependence is more reminiscent of the initial stage of photoionization[2] than the initial stage of resonant excitation of discrete levels. In the latter case, as is well known, there are produced Rabi oscillations[3] whose initial stage corresponds to a quadratic $w(\tau)$ dependence.

In the case of long pulses, when condition (5) is not met, it is impossible to change to integration when w_n (3) is summed. For $\tau\gg n_0^3$, on the contrary, the principal role in the sum $\Sigma_n w_n$ is played by one term $n=n_0$ (or else three, $n=n_0$ and $n_0\pm 1$) with minimum energy-difference modulus $|E_n-E_0-\omega|$. If $|E_n-E_0-\omega|_{\min}=0$, the $w(\tau)$ increases quadratically, corresponding to the initial stage of Rabi oscillations.[3] If, however $|E_n-E_0-\omega|_{\min}\tau\gg 1$, then $w(\tau)$ and all $w_n(\tau)$ decrease exponentially, meaning that the field is turned on and off adiabatically slowly in the nonresonant case. The complete function $w(\tau)$ is shown in Fig. 1a.

The onset of the linear $w(\tau)$ dependence (6) in lieu of the quadratic one during the initial stage [under condition (5)] is not yet a manifestation of some interference effect. It is simply the result of addition of probabilities. The probability w_n (3) of transition to each Rydberg level E_n under condition (5) is proportional to τ^2. The number, in the sum over m, of those terms which make a noticeable contribution to w is of the order of $\tau^{-1}/\Delta_{n_0}=n_0^3\tau^{-1}$, and it is this which leads to the linearity of $\omega(\tau)$ (6). Even this very simple result, however, shows that under condition (5) there is essentially no difference between photoionization and excitation of high Rydberg levels.

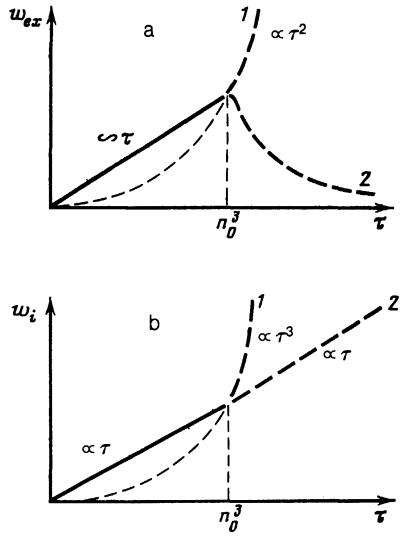

FIG. 1. Probability of excitation (a) and of two-photon ionization (b) vs the pulse duration τ: 1—exact resonance, 2—nonresonant case.

We find now in second-order perturbation theory the probability of two-photon ionization, assuming as before that ω is close to but smaller than $|E_0|$. The probability amplitude of a transition to a continuum state of energy $E\sim E_0+2\omega$ during the entire time of action of the pulse (1) is equal to

$$C_E(\infty)=-i\sum_n\frac{1}{2}V_{En}\int_{-\infty}^{+\infty}dt'\exp\{i(E-E_n-\omega)t'\}C_n(t')f(t'), \tag{7}$$

where the $C_n(t)$ are defined by Eq. (2).

The dependence of the matrix elements V_{En} of the bound-free transitions on n is similar to (4) (Ref. 1):

$$V_{En}=Vn^{-3/2}, \tag{8}$$

where $V=$ const. For $\omega\ll 1$, according to Refs. 4 and 5,

$$V\sim F_0/\omega^{5/2}. \tag{9}$$

Actually, of course, the "constant" V depends on E, but this dependence is slow and can be neglected if there are no autoionizing levels in the energy region $E\sim E_0+2\omega$ (the flat continuum approximation[6]).

Using Eqs. (7)–(9), it is easy to find also the total ionization probability

$$w_i=\int dE|C_E(\infty)|^2$$
$$=\frac{1}{2}\pi V^2\int_{-\infty}^{+\infty}dt f^2(t)\left|\sum_n n^{-3/2}C_n(t)e^{-iE_nt}\right|^2. \tag{10}$$

In the integration with respect to E we have used here the usual approximation with a large excess above the threshold,[2,7] an approximation based on the assumption that the width ($\sim 1/\tau$) of the continuum populated-levels bands is much less than $E_0+2\omega$. This allows us to extend the limits of the integration with respect to E to $[-\infty,+\infty]$.

Equation (10) is a direct generalization of the known

equations that describe resonant ionization of an atom via an isolated resonant level in the regime of broadening of the resonance due to the short duration of the pulse.[7-9]

Under condition (5), it is possible again in Eq. (10) to change in the sum over n to integration with respect to E_n; with allowance for (2) and (4), this yields

$$w_i = \frac{1}{8}\pi^3 V_{10}^2 V^2 \int dt f^4(t) = \frac{1}{16}\pi^{3/2}\tau V_{10}^2 V^2. \qquad (11)$$

On the contrary, for $\tau \gg n_0^3$ and $|E_n - \omega - E_0|_{\min}\tau \ll 1$ (exact resonance), the main contribution in the sum over n in Eq. (10) is made by one single term (with $n = n_0$):

$$w = \frac{\pi V_{10}^2 V^2}{32 n_0^6}\int_{-\infty}^{+\infty} dt f^2(t)\left(\int_{-\infty}^{t} f(t')\,dt'\right)^2 \propto \tau^3. \qquad (12)$$

At $\tau \gg n_0^3$ and $|E_n - \omega - E_0|_{\min}\tau \gg 1$ the sum over n in (10) receives contributions from several terms with n close to n_0, and for these terms

$$C_n(t) \approx -\frac{V_{10} f(t)}{2n_0^{3/2}(E_n - E_0 - \omega)}\exp\{i(E_n - E_0 - \omega)t\} \qquad (13)$$

and

$$w_i = \frac{\pi^3}{8}\frac{V^2}{n_0^6}\int_{-\infty}^{+\infty} dt f^4(t)\left(\sum_n \frac{1}{E_n - E_0 - \omega}\right)^2$$

$$= \frac{\pi^{3/2} V_{10}^2 V^2 \tau}{16 n_0^6}\left(\sum_n \frac{1}{E_n - E_0 - \omega}\right)^2. \qquad (14)$$

The complete function $w_i(\tau)$ is shown in Fig. 1b. The region $\tau \gg n_0^3$ is that of ordinary resonant ionization through an isolated level[2,7-9] [for $|E_n - \omega - E_0|_{\min}\tau \ll 1$ (12) (Fig. 1b, curve 1)] or through direct nonresonant two-photon ionization [for $|E_n - \omega - E_0|_{\min}\tau \gg 1$ (14) (Fig. 1b, curve 2)]. There are no interference effects at all in this region. At exact resonance we have $w_i \propto \tau^3$, as is well known from the theory of resonance ionization.[2,7-9]

The region $\tau < n_0^3$ (5) is the interference region. In this case the probability amplitudes of the excitation of the Rydberg levels $C_n(t)$ are coherently added in the sum (10) over n. The number of terms that make a noticeable contribution to the sum is of the order of $\tau^{-1}/\Delta_{n_0} = n_0^3/\tau$, which increases the ionization probability by n_0^6/τ^2 times and leads to a linear $w_i(\tau)$ dependence (11). The total ionization probability w_i (11) is smaller than the total probability w (6) of excitation of Rydberg levels if $V \ll 1$ [Eq. (9)]. This is the condition for the validity of perturbation theory with respect to the second degree of two-photon ionization under conditions (5) for interference of intermediate Rydberg levels.

3. PHOTOIONIZATION OF COHERENTLY POPULATED RYDBERG LEVELS BY A SECOND RADIATION PULSE

It was assumed up to now that the transitions to the continuum were induced by the same pulse that produced the coherent population of the Rydberg levels. With the problem so formulated, the width of a packet of coherently populated Rydberg levels was uniquely connected with the duration of the interaction. Another formulation of the problem is also possible, as mentioned in the Introduction. One can produce a packet of coherently populated Rydberg levels of definite width ΔE by a single electromagnetic-field pulse, and the photoionization or other transitions from this state can be produced by another pulse of different duration τ. In this formulation of the problem, the connection is lost between the initial width ΔE of the packet and the pulse duration τ.

Thus, let the wave function of the atom be given at the initial instant $t = 0$ in the form of a packet of coherently populated Rydberg levels.

$$\Psi(t=0) = \sum_n C_n^{(0)}\varphi_n. \qquad (15)$$

We normalize the wave function (15) to unity; this yields

$$\sum_n |C_n^{(0)}|^2 = 1. \qquad (16)$$

Strictly speaking, it is necessary to sum in (15) not only over the principal quantum number $n \gg 1$ but also over the orbital quantum number l and its projection m_l. We assume for simplicity that only the expansion coefficients corresponding to some definite $l \ll n$ differ from zero in this sum. Such a situation obtains, for example, in the case of coherent population of Rydberg levels, which occurs in a one-photon transition from the ground S state. In this case all the φ_n in the sum (15) are wave functions of P states.

If Δn is the width of the packet (15), $n \gg l$, and the probability amplitudes $C_n^{(0)}$ are smooth functions of n, i.e., change little when n changes by unity, then it is possible to change over in the sum (16) to integration with respect to n. Taking this circumstance into account, we approximate C_n^0 by a Gaussian function,

$$C_n^{(0)} = \frac{1}{\pi^{1/4}(\Delta n)^{1/2}}\exp\left(-\frac{(n-n_0)^2}{2\Delta n^2}\right), \qquad (17)$$

where n_0 is the principal quantum-number corresponding to the center of the packet $C_n^{(0)}$ ($n_0 = \bar{n}$).

Assume that n_0 and Δn satisfy the conditions

$$n_0 \gg \Delta n \gg 1, \qquad (18)$$

which means that the theoretical packet width $\Delta E = \Delta n \Delta_{n_0} = \Delta n/n_0^3$ is much larger than the distance Δ_{n_0} between the nearest levels, but much smaller than the distance to the continuum threshold $1/2n_0^2$.

In the general case the C_n^0 depend on the method of producing the wave packet. If the packet is produced by the short radiation pulse that excites the atom, then $C_n^{(0)}$ coincides with $C_n(\infty)$ (2), i.e., it is determined by the Fourier transform of the envelope of the exciting field. If this envelope is the Gaussian function (1), then $C_n^{(0)}$ is also described by the Gaussian relation (17). By virtue of the condition (18) the power-law variation of the matrix elements V_{0n} in (2) with n is slow over an interval $\sim\Delta n$; we can therefore neglect this variation and put $V_{0n} \approx V_{0n_0}$. Just as before, all the results obtained below do not depend qualitatively on the explicit form of $C_n^{(0)}$. The Gaussian dependence (18) will be used for illustration and to obtain ultimately correct numerical coefficients.

Let now the atom be acted upon by a pulse of electromagnetic radiation of frequency ω, maximum electric field amplitude F_0, pulse duration τ, delay time t_0 of the field max-

imum relative to the instant $t=0$ of packet formation (15), and temporal envelope

$$f(t)=\exp(-(t-t_0)^2/\tau^2). \tag{19}$$

All the remarks made above concerning the envelope (1) are applicable to the choice of the explicit form of the envelope (19).

The probability amplitudes $C_E(\infty)$ of the transition to the continuum states, calculated in first-order perturbation theory, are determined by the previous equation (7), in which the lower integration limit must be replaced by $t'=0$ and the functions $C_n(t)$ by $C_n^{(0)}$ (17). The function (19) must be used for the envelope $f(t')$.

The total ionization probability is determined by the integral of $|C_E(\infty)|^2$ over the energies E; this integral is calculated in an approximation with a large excess above the threshold.[2,7-9] It is implied that the bandwidth $\sim\max(\tau^{-1}, \Delta E)$ of the populated levels of the continuum is small compared with $E_n+\omega$, where $E_{n_0}=-1/2n_0^2$, so that the limits of integration with respect to E can be extended to $-\infty$ and $+\infty$. The calculation result is

$$w=\frac{\pi V^2}{2n_0^3}\sum_{n,n'} C_n^{(0)}C_{n'}^{(0)}\int_0^\infty f^2(t)\exp\left(i\frac{n-n'}{n^3}t\right)dt, \tag{20}$$

where we used, by virtue of the condition (18), expansions of the energies E_n and E_n' near E_{n_0}. We change in (20) from summation over n and n' to summation over n and $m\equiv n'-n$. After this we replace the sum over n by an integral, something always possible since, by virtue of the conditions (18), the characteristic interval of the values of n, over which the integrand changes substantially, is always greater than unity. As a result of integration with respect to n, with allowance for the explicit form of $C_n^{(0)}$, we obtain from (20)

$$w=\frac{\pi V^2}{2n_0^3}\sum_m\int_0^\infty dt f^2(t)\exp\left\{-\frac{m^2}{4\Delta n^2}-i\frac{mt}{n_0^3}\right\}. \tag{21}$$

In the general case of arbitrary $C_n^{(0)}$, the factor $\exp(-m^2/4\Delta n^2)$ in (21) will be replaced by some function of m which has a at $m=0$ a maximum of width $\sim\Delta n$.

We can change in (21) to integration with respect to m if $t\ll n_0^2$, as is the case when

$$\tau\ll n_0^3, \quad t_0\ll n_0^3. \tag{22}$$

Under these restrictions on τ and t, replacement of the sum by an integral in (21) and integration with respect to m yield

$$w_i=\pi^{1/2}V^2\Delta E\int_0^\infty dt f^2(t)\exp(-(\Delta E)^2t^2)=\frac{\pi^2V^2\Delta E\tau}{2^{3/2}(1+\tau^2(\Delta E)^2/2)^{1/2}}$$
$$\times\exp\left\{-\frac{t_0^2\Delta E^2}{1+\tau^2(\Delta E)^2/2}\right\}\left[1+\Phi\left(\frac{2^{1/2}t_0/\tau}{(1+\tau^2(\Delta E)^2/2)^{1/2}}\right)\right], \tag{23}$$

where $\Phi(x)$ is the probability integral,[10] $\Phi(x)\approx 1$ at $x\gg 1$ and $\Phi(x)\approx 0$ at $x\ll 1$.

For arbitrary $C_n^{(0)}$, the factor $\exp(-(\Delta E)^2t^2)$ in (23) is replaced by the Fourier transform of a slow function of m which replaces $\exp(-m^2/4\Delta n^2)$ in (21), at the frequency tn_0^{-3}.

Depending on t_0, the exponential factor in Eq. (23) describes an exponential decrease, whereas the factor in the square bracket is doubled with increase of t_0. The ratio of these two factors is different at large and small values of the parameter ΔE_τ.

If $\Delta\varepsilon\tau\ll 1$, the time of the exponential decrease of $w_i(t_0)$ (23) is equal to $(\Delta E)^{-1}$, and the time for the probability integral Φ to increase from 0 to 1 is of the order of $\tau\ll(\Delta E)^{-1}$. The transition probability $w_i(t_0)$ first decreases rapidly to half its value within an interval $t_0\sim\tau$; this is followed by a slower exponential decrease with a rate constant equal to ΔE. If, however $\Delta E\tau\gg 1$, the characteristic time $t_0\sim\tau$ of the exponential decrease of $w_i(t_0)$ (23) is much smaller than the growth time of the probability integral from 0 to 1, which is of the order of $\tau^2\Delta E\gg\tau$. In this case the maximum of $w_i(t_0)$ is much less pronounced than at small values of $\Delta E\tau$ (Fig. 2).

The described $w_i(t_0)$ dependence is qualitatively understandable. If the initial packet (15) is ionized at $t_0=0$ by only half a pulse with $t>0$, then at $t_0>\tau$ almost the entire ionizing radiation pulse (that part of the pulse for which $t>0$) takes part in this process. If at the same time $\Delta E\tau\ll 2$, neither noticeable dephasing of the wave functions of the packet (15) nor the associated decrease of the ionization probability takes place within the time τ. Therefore, when the time t_0 varies from 0 to τ the ionization probability increases in proportion to the area under the $f^2(t)$ curve with $t>0$, i.e., by an approximate factor of two. If, however, $\Delta E\tau\gg 1$, this effect is suppressed by the rapid decrease [within a time $\sim(\Delta E)^{-1}$] of the ionization probability, owing to the dephasing of the terms in the sum (15) at $t>0$. It is clear thus that the form of the $w_i(t_0)$ curves in Fig. 2 does not depend qualitatively on the explicit forms of $C_m^{(0)}$ and $f(t)$.

The described behavior of the dependence of the ionization probability on the delay time t_0 takes place only at small values of t_0, determined by the second inequality of (22). For longer t_0, $t_0\gtrsim n_0^3$, a change to integration in the sum (21) over m is generally speaking impossible. Let us consider, however, values of t_0 that are close to $2\pi kn_0^3$, where $k=1, 2, \ldots$, i.e., let

$$t_0=2\pi kn_0^3+t_0', \tag{24}$$

where t_0' satisfies the second condition in (22), $t_0'\ll n_0^3$. In this case replacement of the sum over m by an integral is possible, and leads to Eq. (23) with t_0 replaced by t_0', and with shifted values of the lower limit of the integral with

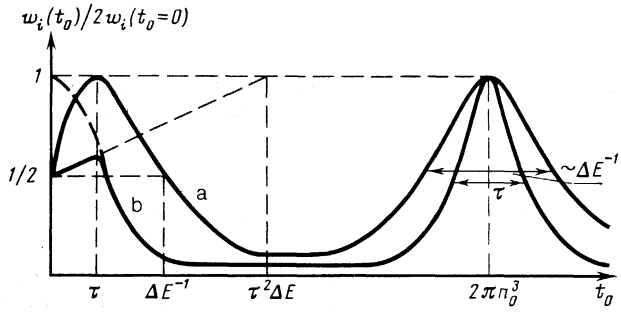

FIG. 2. Dependence of the ionization probability on the delay time t_0 at $\Delta E\tau\ll 1$ (a) and $\Delta E\tau\gg 1$ (b).

respect to t and of the argument of the probability integral Φ. The latter, as can be easily verified, takes the form

$$\Phi\left(\frac{2^{1/2}t_0/\tau}{(1+\Delta E^2\tau^2/2)^{1/2}}+\frac{2^{3/2}\pi k n_0^3}{\tau}(1+\Delta E^2\tau^2/2)^{1/2}\right)\approx 1. \tag{25}$$

Since the first of the conditions (22) is assumed met, the argument of the probability integral in (25) is large and $\Phi\approx 1$ for all $k\geqslant 1$. The function $w_i(t_0)$ is periodic starting with $t_0=2\pi n_0^3$, and its period is $2\pi n_0^3=2\pi/\Delta_{n_0}$. Regardless of the structure of the first maximum of $w_i(t_0)$ (at small t), $w_i(t_0=2\pi k n_0^3)=2w_i(t_0=0)$ in all the succeeding maxima (Fig. 2).

The reason for the twofold increase of $w_i(2\pi k n_0^3)$ over $w_i(t_0=0)$ is that at $k\neq 0$ the ionization is effected during the entire pulse $f(t)$ and not during half the pulse as in the case $k=0$.

The periodic character of the function $w(t)$ has a simple physical cause. The wave packet (15) describes the state of an electron localized near the nucleus of the atom. The electron usually moves away from the nucleus with time along an elongated elliptic orbit. Its interaction with the nucleus weakens, it becomes more and more free, and its ability to absorb a photon decreases (a completely free electron cannot absorb a photon). This process is described by the exponential decrease of $w_i(t_0)$ in accordance with (23) (Fig. 2). After a time $2\pi/\Delta_{n_0}=2\pi n_0^3$, however, the electron again approaches the nucleus and the photoionization probability again increases. Taken by itself, the periodic increase of $w_i(t_0)$, without analysis of the structure of the first maximum of $w_i(t_0)$, was described by another method in a recent paper.[11]

We proceed now to analyze the dependence of the probability $w_i(\tau)$ on the pulse duration. We consider the simplest case, when the delay time t_0 is short enough

$$t_0\ll\tau(1+\Delta E^2\tau^2/2)^{1/2},\quad (\Delta E)^{-1}(1+\Delta E^2\tau^2/2)^{1/2}. \tag{26}$$

Under these restrictions on t_0, Eq. (23) assumes the simpler form

$$w_i=\frac{\pi^2}{2^{3/2}}\frac{V^2\Delta E\tau}{(1+\Delta E^2\tau^2/2)^{1/2}}. \tag{27}$$

For large and small τ ($\tau\Delta E\ll 1$ and $\tau\Delta E\gg 1$), $w_i(\tau)$ is respectively a linear function and a constant

$$w_i=\frac{\pi^2}{2^{3/2}}V^2\Delta E\tau,\quad \Delta E\tau\ll 1,$$

$$w_i=\frac{\pi^2}{2}V^2,\quad \Delta E\tau\gg 1. \tag{28}$$

In the general case of arbitrary $C_n^{(0)}$ and $f(t)$ it is impossible to obtain a simple formula of type (27), but qualitatively all the relations in (28) remain in force. When $C_n^{(0)}$ and $f(t)$ deviate from the Gaussian curves (17) and (19), only the numerical coefficients in Eqs. (28) are changed.

Equations (27) and (28), however, are valid only so long as τ is not too large [the first of the restrictions in (22)]. For larger τ ($\tau\gg n_0^3$) it is necessary to retain in the sum (21) over m only the term with $m=0$; this yields

$$w_i=\frac{\pi^2V^2}{2n_0^3}\int_0^\infty f^2(t)\,dt$$

$$=\frac{2^{1/2}\pi^{3/2}}{4}V^2\tau\Delta_{n_0},\quad \tau\gg\Delta_{n_0}^{-1},\ t_0. \tag{29}$$

Consequently, at large τ the linearity of the photoionization probability in the pulse duration τ is restored, but the growth rate of w_i is in this case much smaller than for small τ (28):

$$w_i\Big|_{\substack{\tau\ll\Delta E^{-1}\\ t_0=0}}=\frac{\pi^2V^2\Delta E\tau}{2^{3/2}}\sim\Delta n w_i\Big|_{\substack{\tau\gg\Delta_0^{-1}\\ t_0=0}}. \tag{30}$$

The $w_i(\tau)$ dependence is totally represented in Fig. 3a. The effect of the initial rapid growth of $w_i(\tau)$ and of its subsequent leveling off to form a plateau are due to interference between different ionization channels. In fact, we can regard the ionization as an independent transition of each of the populated discrete levels to the continuum. The width of the maximum of the photoelectron energy distribution due to each such transfer is $1/\tau$. If $1/\tau\gg\Delta_{n_0}$, the neighboring maxima overlap and they can interfere. If $1/\tau\gg\Delta E$, all the maxima whose amplitudes are not small overlap. This is the case of maximum interference. It is seen directly from (20) that in this case

$$w_i\propto\left(\sum_n C_n^{(0)}\right)^2, \tag{31}$$

which confirms the total-interference conclusion, since it is obvious that what are added in (31) are probability amplitudes and not probabilities $|C_n^{(0)}|^2$ of population of individual levels.

The probability of a transition to the continuum from each of the Rydberg levels has an order of magnitude $V^2\Delta_{n_0}\tau$. The number of such transitions [the number of terms in the sum (31) over n] is of the order of Δn, which leads to an increase of the ionization probability by $(\Delta n)^2$ times.

Finally, the probability of finding an atom on each of the Rydberg levels is $1/\Delta n$, which explains qualitatively in final analysis the increase of the total ionization probability by Δn times (30).

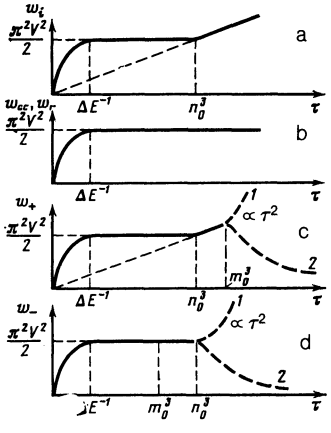

FIG. 3. Probabilities of photoionization (a), of free-free transitions or of induced recombination (b), and of bound-bound transitions "upward" [$m_0>n_0$, (c)] and "downward" [$m_0<n_0$, (d)] as functions of the pulse duration τ.

The case $1/\tau \gg \Delta E$ is the limit of the manifestation of the difference between coherent and noncoherent population of discrete levels.

If $\Delta E \gg 1/\tau \gg \Delta_{n_0}$, there is incomplete overlap of all the maxima produced in the continuum upon ionization of effectively populated discrete levels. Consequently, the interference in this case is likewise incomplete. The probability of transition from each level is as before $V^2\Delta_{n_0}\tau$, the number of interfering maxima becomes $\sim 1/\tau$ in this case, so that the total probability $\sim V^2\Delta_{n_0}$ is independent of τ. This explains the plateau on Fig. 3a.

Finally, at $1/\tau < \Delta_{n_0}$ the maxima in the distribution of the photoelectrons become so narrow that they do not overlap at all. In this case there is no interference. The photoionization probability is determined by Eq. (29). It is obvious from (20) that in this case

$$w_i \infty \sum_n |C_n^{(0)}|^2. \tag{32}$$

Since there is no interference, there is likewise no difference between the coherent and noncoherent population of the levels: the photoionization probability depends only on the squares of the modulus of the probability amplitudes $C_n^{(0)}$.

We point out that at $\tau \gg n_0^3$ the conclusion that there are no interference effects is valid independently of the width of the wave packet (15). Even in the case $\Delta n \gg 1$, $\Delta E \gg n_0^3$, when the packet is broad and includes a large number of Rydberg levels, there is no interference at $\tau \gg n_0^3$, i.e., the photoionization is produced just as if the electron were on one isolated level E_{n_0}. This means that coherent population of a large number of levels is not enough for interference to be present; it is necessary also that the duration of the pulse that causes the transition to the continuum be short enough (5).

We note, finally, that the criterion for the validity of the perturbation theory expounded above is smallness of the ionization probability (28), $w_i \ll 1$. The condition that w_i be small in the vicinity of the plateau on Fig. 3a has the same form $V \ll 1$ as before.

4. TRANSITIONS TO THE CONTINUUM AND INDUCED RECOMBINATION

It was assumed up to now that the initial packet (15) is produced in the region of highly excited discrete levels. A different situation is also possible, when the initial packet is produced in the continuum

$$\Psi(t=0) = \int dE C_E^{(0)} \varphi_E,$$

$$C_E^{(0)} = \frac{1}{\pi^{1/4}(\Delta E)^{1/2}} \exp\left(-\frac{(E-\bar{E})^2}{2(\Delta E)^2}\right), \tag{33}$$

where φ_E are the atomic functions of the continuous spectrum, normalized to an energy δ-function, and $\Psi(t=0)$ is normalized to unity.

Just as above, the explicit form of the function $C_E^{(0)}$ is specified by the Gaussian curve (33). This enables us to carry out all the calculations through to conclusion, and determine not only the principal relations, but also the numerical coefficients. The latter vary with the form of $C_E^{(0)}$ or $f(t)$, but all the qualitative relations are independent of the explicit forms of $C_E^{(0)}$ and $f(t)$.

If $\omega < \bar{E}$, the electromagnetic-radiation pulse (19) can produce only transitions from the state (33) into other continuum states $E \to E \pm \omega$. If $\omega > \bar{E}$, induced recombination $E \to E_n \sim E - \omega$ is also possible. We emphasize that in contrast to the usually considered processes,[12] in this case we are dealing with induced recombination on an individual atom, namely on the particular atom whose ionization by the preceding pulse produced the photoelectrons.

The matrix elements of the bound-free transitions are determined by Eqs. (8) and (9). The matrix elements $V_{EE'}$ of free-free transitions are determined, in the approximation of a flat continuum[6] by the same constant V [Eq. (9)], Refs. 4 and 5:

$$V_{EE'} \approx V = \text{const}.$$

In both considered cases (free-free transitions and induced recombination) the result of the calculation of the transition probability w reduces to the foregoing (23), but without the restrictions (22) on the pulse duration τ and on the delay time t_0. This is understandable: the electron in the continuum moves away from the atom and does not return to its initial position. Depending on the pulse duration τ, no region of repeated slow linear growth appears in the transition probabilities (Fig. 3b). This means that upon formation of a wave packet in the continuum and in the succeeding transitions (disregarding the influence of the neighboring atoms), a definite degree of interference remains at all τ, and nothing imposes an upper bound on the plateau region in Fig. 3b.

In the case of free-free transitions in an isolated atom, the laws formulated are almost obvious. They follow from the analysis of preceding section, in which n_0 should be made to tend to infinity, thereby lifting the restrictions (22) on t_0 and τ.

Let us dwell in somewhat greater detail on the case of induced recombination. The probability amplitudes of transitions from state (33) into states φ_n corresponding to Rydberg levels E_n can be easily found in first-order perturbation theory:

$$C_n(\infty) = -\frac{i}{2}\int dE C_E^{(0)} V_{nE} \int_0^\infty dt f(t) \exp\{i(E_n - \bar{E} + \omega)t\}. \tag{34}$$

Substituting $C_E^{(0)}$ of (33) in (34) and using expressions (8) and (9) for the matrix elements V_{nE}, we find that the probability w_r of the induced recombination can be represented in the form

$$w_r = \sum_n |C_n(t=\infty)|^2 = \pi^{1/2} V^2 \int_0^\infty dt \int_0^\infty dt' f(t) f(t'),$$

$$\sum_n n^{-3} \exp\{i(E_n - \bar{E} + \omega)(t-t') - {}^1/_2 (\Delta E)^2 (t^2 + t'^2)\}. \tag{35}$$

As a function of each of the variables t and t', the integrand of (35) has maxima at $t, t' = 0$ and $t, t' = t_0$, with respective widths $\sim\tau$ and $\sim(\Delta E)^{-1}$. At small t_0 the maxima are equal and have a width $\sim \min(\tau, (\Delta E)^{-1})$. The same quantity limits $|t - t'|$ from above:

$$|t-t'| \lesssim \min\{\tau, (\Delta E)^{-1}\}. \quad (36)$$

It follows hence that it is possible to change in the sum (35) over n to integration with respect to E_n if

$$\Delta_{n_0} \equiv \frac{1}{n_0^3} \ll \max\left\{\frac{1}{\tau}, \Delta E\right\}, \quad (37)$$

where Δ_{n_0} is the distance between the nearest Rydberg levels in the resonance region with initial energy $\sim \bar{E}$, $E_{n_0} = -(2n_0^2)^{-1} \approx \bar{E} - \omega$. It follows from (37) that the sufficient condition for the change from summation to integration in Eq. (35) is smallness of Δ_{n_0} compared with the initial width ΔE of the packet in the continuum, independently of the pulse duration τ. Assuming that $\Delta_{n_0} \ll \Delta E$ and changing in (35) to integration with respect to E_n we obtain as a result expression (23) for the probability of the induced recombination. Consequently, at $\Delta_{n_0} \ll \Delta E$ transitions from the continuum to highly excited Rydberg levels do not differ from the transition in the continuum. All the conclusions concerning the functions $w(t_0, \tau)$, formulated above and illustrated in Fig. 3b, are also valid in this case.

5. TRANSITIONS BETWEEN GROUPS OF RYDBERG LEVELS

We turn again to the formulation of the problem described in Sec. 3, in which a packet (15) is formed at the initial instant of time $t = 0$ in the region of highly excited Rydberg levels.

Let now the frequency ω be less than the distance from the center of the packet, $E_{n_0} = -1/2n_0^2$, to the threshold of the vacuum, $\omega < 1/2n_0^2$. This raises the question of transitions between two groups of closely located levels with a distance $\sim\omega$ between the groups of levels.

Let the index m label the levels of the initially non-populated group of levels, to which the transitions take place.

According to Refs. 4 and 5, at low values of the orbital momentum $l \ll m$, n and m, $n \gg 1$, the matrix elements V_{mn} can be represented in the form

$$V_{mn} = V/(mn)^{3/2}, \quad (38)$$

where V is the same constant (9) as in the matrix elements of the free-free and bound-free transitions.

The total probability of the transition from the level group (n) to the level group (m) is obtained in analogy with the foregoing:

$$w = \frac{1}{4} V^2 \sum_{n,n'} C_n^{(0)} C_{n'}^{(0)} (nn')^{-3/2} \int_0^\infty dt \int_0^\infty dt' f(t) f(t') \sum_m m^{-3} \exp\{iE_m(t-t') - i(E_n \pm \omega)t + i(E_{n'} \pm \omega)t'\}, \quad (39)$$

where the $\pm$ signs correspond to "up" and "down" transitions, for which $m > n$ and $m < n$, respectively.

If the pulse duration is not too long

$$\tau < m_0^3, \quad (40)$$

where m_0 is the center of the excited-levels band $((2m_0^2)^{-1} \approx (2n_0^2)^{-1} \mp \omega)$, the summation over m can be replaced by integration. In the expression obtained, the preexponential factor $(nn')^{-3/2}$ can be replaced by n_0^{-3}, and the energies E_n and $E_{n'}$ can be located near E_{n_0}, so that the probability w can be represented in the form (23). Consequently, if condition (40) is met all the conclusions of Sec. 3 concerning the dependence of the probability w on t_0 and τ are valid. This means that under condition (40) the excited group of discrete levels does not differ essentially from the continuum. If $m_0 > n_0$, the condition (40) is less stringent than the first of the conditions (22). In this case one can track, within the framework of condition (40), all the stages, illustrated in Fig. 3a, of the variation of the $w(\tau)$ dependence. If the pulse duration at $m_0 > n_0$ become so long that condition (40) is violated, it is necessary to retain in the sums over m, n, and n' (39) only one term corresponding to the minimum value of $|E_n - E_m + \omega|$. The subsequent behavior of the function $w(\tau)$ depends on the relation between τ^{-1} and $|E_n - E_m + \omega|_{\min}$. If $|E_n - E_m + \omega|_{\min}\tau \ll 1$, certain two levels are in exact resonance, one each from the initial and final groups of levels. The probability $w(\tau)$ increases quadratically (Fig. 3c, curve 1).

If $|E_n - E_m + \omega|_{\min}\tau \gg 1$, however, the probability $w(\tau)$ decreases exponentially with increase of τ (Fig. 3c, curve 2).

The reason why $w(\tau)$ decreases in the nonresonant case is that a transition to the adiabatic limit takes place with increase of τ.

If $m_0 < n_0$, the condition (40) is violated earlier than the first of the conditions (22). There exists a range of τ values

$$(\Delta E)^{-1} \ll m_0^3 < \tau < n_0^3, \quad (41)$$

in which the foregoing conclusion is not valid. In this case it is necessary first to sum over n and n' in (39), using the explicit expression (17) for $C_n^{(0)}$ and replacing these sums by integrals. The resultant sum over m can, by virtue of the condition $m_0^3 \gg (\Delta E)^{-1}$, also be replaced by an integral that can be easily calculated and leads to the previous result (23), according to which $(\Delta E)^{-1} w = \text{const}$ for $\tau \gg t_0$ all the way to $\tau \sim n_0^3$.

For $\tau > n_0^3$, $n_0 > m_0$ it is again impossible to replace the sums over n and n' in (39) with an integral. On the contrary, it is necessary to retain in the sums only one term with the minimum values of $|E_n - E_m - \omega|$ and $|E_{n'} - E_m - \omega|$. Depending on the relation between $|E_n - E_m - \omega|_{\min}$ and $1/\tau$, the transition probability either increases quadratically or decreases exponentially (Fig. 3d). Consequently, the only difference between the cases $n_0 < m_0$ and $n_0 > m_0$ (Figs. 3c and 3d) is that in the former case the $w(\tau)$ curve has a section $n_0^3 < \tau < m_0^3$ with a second slow linear growth of (29), and in the second case there is no such section.

6. CONCLUSION

Let us formulate the main conclusions that follow from the foregoing analysis.

1. At a fixed duration of the pulse that causes the transitions between Rydberg levels, there exists a certain critical level number $n_c = \tau^{1/3}$ above which (at $n > n_c$) there is in fact no difference between transitions in a system of Rydberg levels and transitions in a continuum. Below this critical n ($n < n_c$), on the contrary, there is no interference whatever between transitions in a continuum or in a system of high Rydberg levels. When it comes to transitions between discrete-spectrum states, transitions at $n < n_c$ take place only between resonant level pairs, notwithstanding the possibility

of coherent initial population of a large number of levels $\Delta n \gg 1$. As for photoionization, its probability at $n < n_c$ is proportional to τ and to the number of particles in the band of initially populated levels (32).

2. In both a continuum and in a discrete spectrum interference exists above a critical value of n ($n > n_c$), increases the transition probabilities, makes them dependent on the width ΔE of the packet of coherently populated levels, and leads to a characteristic dependence on the pulse duration. For $\Delta E\tau < 1$ the interference is a maximum and $w \propto \tau$ for $E = \text{const}$, while for $\Delta E\tau > 1$ we have partial interference and $w = \text{const}$.

3. The entire foregoing analysis was based on the use of perturbation theory. Generalization to the case of stronger fields deserves an independent investigation. Since free-free, bound-free, and bound-bound transitions proceed at $n > n_c$ essentially in like manner and are determined by one and the same constant V (11), we can formulate for the validity of perturbation theory the one criterion $V \ll 1$ or

$$F_0^2 \ll \omega^{10/3}. \tag{42}$$

A similar criterion was obtained in Ref. 13.

Note, however that in even weaker fields the dynamic Stark effect can lead to a substantial restructuring of the levels. Under nonresonance conditions, the Stark shift of high levels, just as the shift of the ionization threshold, is equal to the free-electron oscillation energy $F_0^2/4\omega^2$ in the wave field.[14] This shift, which is the same for all levels, does not influence the dynamics of any of the processes. Corrections to $F_0^2/4\omega^2$ are responsible for the level restructuring and can possibly not be small even in relatively weak fields. Particularly significant is the level coupling due to the dynamic Stark effect under resonance conditions. It is quite difficult to formulate some single conditions under which the field is weak enough to be able to neglect all the indicated conditions. It is clear, however, that the level restructuring, regardless of its character, does not alter qualitatively the results formulated above. Changes can take place in the characteristic distance between levels or in the instant after which the discrete level structure appears in bound-bound transitions. There is apparently no change, however, in the plateau-like shape of the $w(\tau)$ curves shown in Fig. 3. This is in fact one of our main results. It manifests itself to the fullest extent in the case of transitions in the continuum.

It appears that the Stark effect imposes no restrictions at all in the continuum, since the only meaningful energy shift for the case of a free electron is the oscillation-energy shift.

The transition probabilities in a continuum, according to our results above, increase linearly only in the section $t < (\Delta E)^{-1}$. The plateau region, which begins at $t \sim (\Delta E)^{-1}$, is not bounded from above. At such arbitrarily large t, interference takes place and its degree is such that $w = \text{const}$. Consequently, for a single atom in a continuum, the field-induced transitions and the energy acquisition by the electron can take place only in a limited time interval $t < (\Delta E)^{-1}$. In this region the average electron energy increases linearly with t. At $t > (\Delta E)^{-1}$ no more energy is acquired. It follows hence, in particular, that in a genuine continuum there is no diffusion in energy for a single atom. These statements were proved above only in the weak-field approximation. It follows from the theory of above-threshold ionization,[6] however, that they are valid also in a strong field. For above-threshold ionization ΔE is equal to the multiphoton ionization width Γ_i of the ground level. At $t > \Gamma_i^{-1}$ a stationary distribution is established in the continuum and the atom energy does not increase. Using the results of Ref. 6, the average energy that the electron manages to acquire within a time Γ_i^{-1} can be estimated to be equal to $V\omega$. The interference interpretation presented above reveals the physical cause of the onset of a stationary regime in above-threshold ionization at $t > \Gamma_i^{-1}$.

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