Tunneling ionization from a short-range potential in an intense laser field. Numerical modeling

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Direct numerical integration of the time-dependent Schrödinger equation is used to study the photodetachment of an electron from a short-range potential in the tunneling limit. Dependences of the ionization probabilities on intensity, frequency, and duration of the laser pulse are obtained. The regions of applicability of tunneling and multiquantum ionization mechanisms are determined. The calculated results are compared with analytic models.

1. INTRODUCTION

Nonlinear ionization of atoms in intense light fields has long held the fixed attention of theoreticians and experimentalists.[1–3] The first effort at a general treatment of the ionization of atoms by laser radiation as a transition between the initial state of a discrete and finite continuous spectrum of a quantum system in the field of an electromagnetic wave was undertaken by Keldysh.[4] He showed that the character of the process is determined by the magnitude of the parameter γ (the Keldysh parameter):

$$\gamma=\sqrt{2mI}\,\omega/eE, \tag{1}$$

where E and ω are the electric field intensity and frequency of the electromagnetic wave, and I is the ionization potential of the quantum system. Here the case $\gamma\ll1$ corresponds to the multiquantum ionization regime, and the case $\gamma\gg1$, to the tunneling ionization regime. The quasistatic model of tunneling ionization in a variable field for the hydrogen atom was considered in Ref. 5 and generalized in Ref. 6 to the case of an arbitrary atom. However, until recently essentially all of the experimental data have corresponded to the multiquantum limiting case. The creation of powerful pulsed infrared CO_2 lasers has made experimental study of tunneling ionization possible.[7] Recent experiments (see, e.g., Refs. 8–10) have demonstrated a wide range of applicability of the quasistatic model.

Direct numerical integration of the time-dependent Schrödinger equation for a quantum system in the field of an electromagnetic wave in the one- or two-dimensional approximation has of late become one of the main methods of investigation of the dynamics of a quantum system in an intense optical field. Thus, numerical experiments have been performed to study superthreshold ionization,[11,12] the stabilization of atoms and negative ions in superstrong fields,[13–15] the generation of optical harmonics,[3] induced bremsstrahlung,[16,17] and other processes. However, carrying out numerical modeling in the region $\gamma\leq1$ also turns out to be an extremely complicated problem in comparison with the study of the opposite case $\gamma\gg1$. The reason for this is the large growth in the time required to integrate the Schrödinger equation when modeling the interaction of a pulse of infrared radiation with an atom in comparison with radiation in the visible or ultraviolet, which is what is commonly used in the experiments, both real and numerical. This large growth in the integration time is due to the necessity of considering significantly longer durations of laser action. This also leads to a significant growth in the region of spatial localization of the wave function, which requires an increase in the number of points of the spatial grid and the calculation time. Calculations in the region $\gamma\leq1$ are also possible for visible and ultraviolet radiation, but in this case ionization takes place in the regime of over-the-barrier disintegration of the atom, or at moderate intensities the tunneling probability turns out to be negligibly small.

The present paper is devoted to a study of photodetachment of an electron from a short-range potential by a femtosecond laser pulse in the parameter range corresponding to $\gamma<1$. Dependences of the photodetachment probability on the intensity, frequency, and duration of action are obtained. The range of applicability of the quasistatic model is determined. The transition from the tunneling limit to the multiphoton limit is considered, and a comparison with the Keldysh photoionization theory is carried out.

2. THEORETICAL MODEL

The one-dimensional atomic potential $V(x)$ was chosen to be a rectangular potential well of finite depth:

$$V(x)=\begin{cases}-V_0, & |x|\leq d/2\\ 0, & |x|>d/2\end{cases}. \tag{2}$$

In the calculations we set $V_0=6$ eV and $d=4$ Å. For the indicated parameter values of the potential well there exist two bound states with energies (measured from the boundary of the continuum) $\varepsilon_1\cong-4.82$ eV and $\varepsilon_2\cong-1.68$ eV, characterized by the wave functions $\varphi_1(x)$ and $\varphi_2(x)$ and satisfying the equation

$$H_0\varphi_1=\varepsilon_1\varphi_1,$$

where H_0 is the atomic Hamiltonian.

The interaction with the field of an electromagnetic wave is described in the dipole approximation

$$V(x,t)=-exE(t),$$

where $E(t)$ is the electric field of the wave. We assumed that the laser pulse is Gaussian in shape

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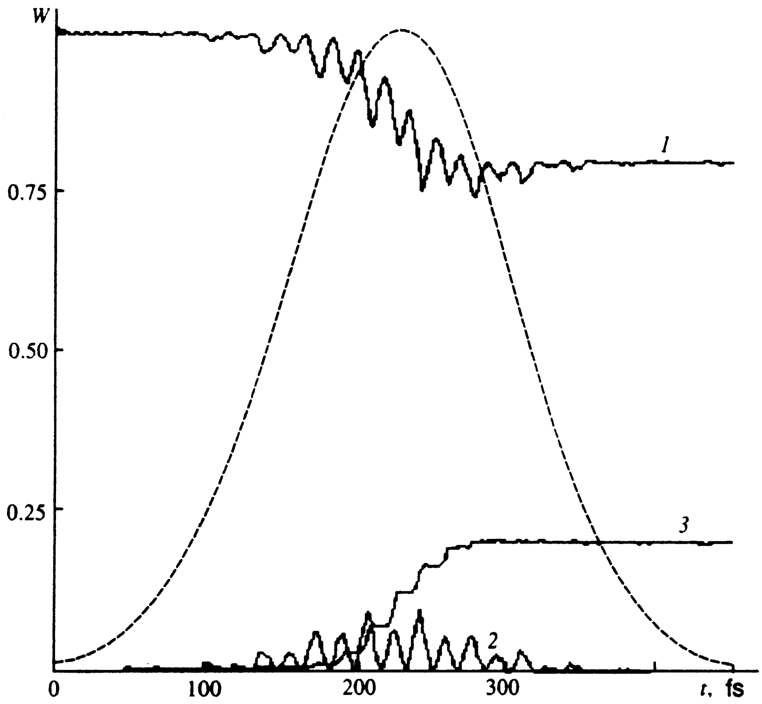

FIG. 1. Time dependence of the probability of detecting an electron in the ground state (*1*), an excited state (*2*), and in the continuous spectrum (*3*) for irradiation by a CO_2-laser pulse of duration 150 fs with maximum intensity 10 TW/cm^2. The dashed line is the envelope of the laser pulse.

$$E(t)=E_0 \exp\left\{-\frac{1}{2}\left(\frac{t-t_0}{\tau}\right)\right\}^2 \cos \omega t, \tag{3}$$

where $\tau_p=2\tau$ is the duration of the laser pulse and t_0 is the time at which the amplitude of the field reaches its maximum value E_0. The energy of a laser photon in the calculations varied from 0.12 eV (CO_2 laser) to 2.34 eV (the second harmonic of a Nd-laser). The duration of the pulse varied from 150 to 400 fs and in all cases satisfied the condition $\omega\tau_p \gg 1$.

Up until the onset of laser action it is assumed that the system exists in the stationary ground state characterized by the wave function $\varphi_1(x)$.

The method of numerically integrating the Schrödinger equation used in the present work is described in Ref. 18. The partitioning of the integration region of dimension $L\sim700$ Å was chosen to be nonuniform so that the greatest numerical accuracy is achieved in the vicinity of the atomic potential. Near the boundaries of the spatial grid an imaginary term is introduced into the potential which causes the wave function to decay and ensures the absence of reflection from the boundaries. This allowed us to study the dynamics of ionization at times greater than the time of movement of a free electron within the limits of the spatial grid.

3. MODELING RESULTS

In the above-indicated range of variation of the frequency of the laser radiation, both the multiphoton and tunneling limits are easily achieved. In the latter case, at radiation intensities $P\leqslant 3\cdot 10^{13}$ W/cm^2 we are talking specifically about tunneling, not about over-the-barrier disintegration of the atom. In a static field, the ionization probability per unit time is given by[5]

$$w=\alpha \exp\left(-\frac{4}{3}\frac{E_a}{E}\right), \tag{4}$$

where $E_a=(2mI^3)^{1/2}/e\hbar$ is the characteristic intra-atomic field, α is some constant, and $I=|\varepsilon_1|$ is the ionization potential.

To calculate the ionization probability in the varying field of an electromagnetic wave, expression (4) must be integrated over time, taking Eq. (3) into account:

$$W=\int_{-\infty}^{\infty} \alpha \exp\left(-\frac{4}{3}\frac{E_0}{E(t)}\right)dt. \tag{5}$$

Assuming that the ionization takes place near the maximum of the electric field and that the condition $\omega\tau_p \gg 1$ is fulfilled, integrating by the method of steepest descent we obtain

$$W=\alpha \frac{3E_0}{2E_a} \exp\left(-\frac{4}{3}\frac{E_a}{E_0}\right)\tau_p. \tag{6}$$

Here $E_0 = \sqrt{8\pi P_0/c}$, where P_0 is the maximum radiation intensity in the pulse.

In the calculations the quantity W was determined from the wave function of the system as follows:

$$W=1-\sum_{i=1,2} |c_i|^2, \tag{7}$$

where

$$c_i(t)=\int \psi(x,t)\varphi_i(x)\exp\left(\frac{i}{\hbar}\varepsilon_i t\right)dx$$

is the amplitude of the probability of detecting an electron at time t in one of the stationary states of the atomic Hamiltonian characterized by energy ε_1 and wave function $\varphi_1(x)$.

Figure 1 presents the dependence of the probability of detecting an electron in the states of the discrete and continuous spectra on time in the radiation field of a CO_2 laser ($\hbar\omega=0.12$ eV) with intensity $P_0=10^{13}$ W/cm^2 and duration $\tau_p=150$ fs. These curves show that ionization of the system

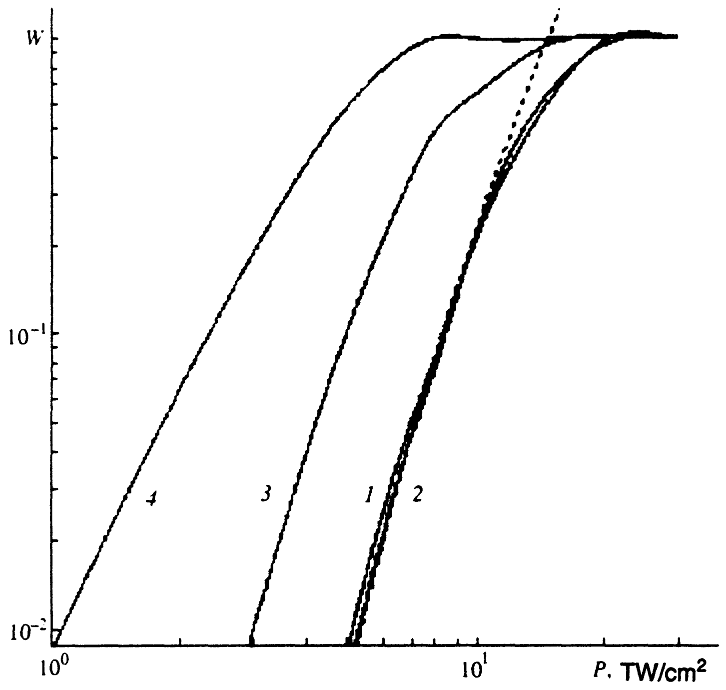

FIG. 2. Dependence of the photoionization probability on the radiation intensity for photon energy $\hbar\omega$ (eV)=0.12 (*1*), 0.24 (*2*), 1.17 (*3*), 2.34 (*4*). The dashed curve represents calculations based on formula (6).

actually takes place near the maximum of the laser pulse at the time when the field intensity of the wave is near its peak value.

It is interesting to note that during the laser pulse there takes place a periodic population of the excited state belonging to the discrete spectrum and that at the end of the laser pulse the population of this state turns out to be zero. Such behavior of a quantum system is associated with adiabatically slow variation of the external electric field during a laser period $T=2\pi/\omega$. Indeed, under the indicated conditions $\omega \ll \omega_{21}=(\varepsilon_2-\varepsilon_1)/\hbar$. Therefore transitions between the states φ_1 and φ_2 of the atomic Hamiltonian can also be treated as the existence of the electron in the ground state $\phi(x,\tau)$ belonging to the system "atom+electric field":

$$(H_0-exE(t))\phi(x,t)=\varepsilon_E\phi(x,t), \tag{8}$$

and the time dependence is parametric.

The validity of the above assertion is confirmed by an estimate of the Rabi frequency $\hbar\Omega_n \simeq (edE)^n/(\hbar\omega)^{n-1}$ if the condition of n-photon resonance is fulfilled:

$$n\hbar\omega=\varepsilon_2-\varepsilon_1. \tag{9}$$

Thus for CO_2 laser radiation with intensity $P\simeq 10^{13}$ W/cm^2 we have $n\simeq 25$, therefore $T=2\pi/\Omega_n\simeq 10^{-12}$ s, which exceeds the duration of the laser pulse.

As can be seen from Eq. (6), in the tunneling limit the quantity W does not depend on the frequency of the radiation. The dependence $W(P_0)$ for different values of $\hbar\omega$, as

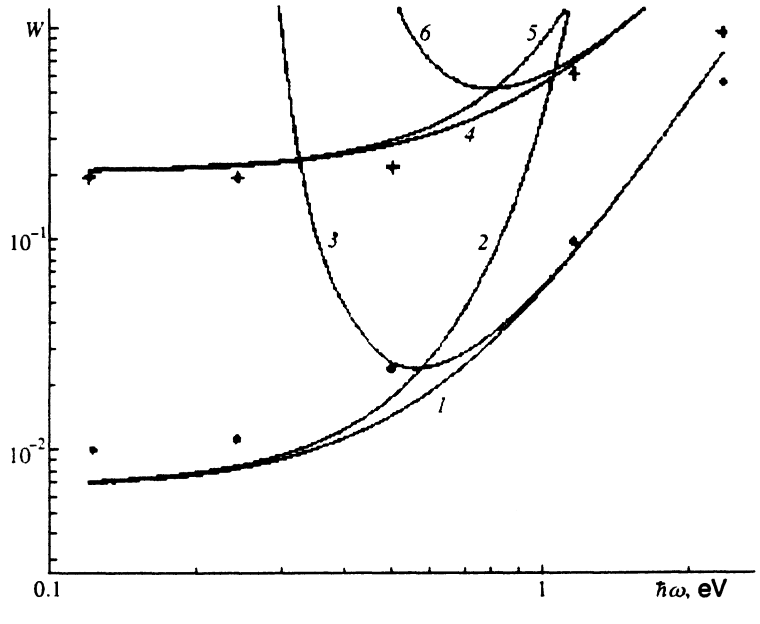

FIG. 3. Dependence of the probability of ionization by a laser pulse of duration 150 fs on the photon energy for P_0=5 TW/cm^2 (points) and P_0=10 TW/cm^2 (crosses). Curves *1* and *4*—calculation according to Keldysh theory (11); curves *2* and *5*—calculation according to formula (12); curves *3* and *6*—calculation according to formula (14).

obtained by numerical modeling, is shown in Fig. 2. The curves corresponding to $\hbar\omega=0.12$ eV and $\hbar\omega=0.24$ eV coincide, and in the region $P_0 \leqslant 1.5 \cdot 10^{13}$ W/cm^2 are well described by relation (6). In the region $P_0 \geqslant 1.5 \cdot 10^{13}$ W/cm^2 saturation is observed in the "experimental" dependence: the ionization probability approaches unity. For $\hbar\omega=1.17$ eV and 2.34 eV (for $P_0=1\cdot 10^{13}$ W/cm^2 the values of the Keldysh parameter γ are approximately 1.6 and 3.3, respectively) the static field approximation is no longer valid. For radiation from a Nd-laser and its second harmonic, the minimum number of absorbed photons required for ionization ($n_{\min}$) is equal, respectively, to 5 and 3, and the logarithm of the ionization probability depends linearly on the logarithm of the intensity in the limit of the multiphoton photoelectric effect:

$$\ln W \sim n_{\min} \ln P_0 . \tag{10}$$

Such a dependence in the region $P_0 \leqslant 5 \cdot 10^{12}$ W/cm^2 (in the absence of saturation) is indeed realized for $\hbar\omega=1.17$ eV and 2.34 eV. However, it turns out that the proportionality constant between $\ln W$ and $\ln P_0$ is equal to 4.2 and 2.6, respectively, for radiation of a Nd laser and its second harmonic, which is somewhat less than expected from the theoretical values.

It is possible to improve the agreement between theory and numerical models in the ionization saturation region if we replace the quantity W in expressions (6) and (10) with the expression $1-\exp(-W)$.

4. NUMERICAL MODELING AND THE KELDYSH IONIZATION THEORY

It would be of especial interest to compare the results obtained with Keldysh theory.[4] This theory assumes that under the action of the wave field, a transition takes place from the initial unperturbed state of the discrete spectrum to a final state of the continuous spectrum, described by the Volkov wave function. For atoms, as a consequence of the long-range nature of their potential, the latter approximation requires additional grounding. The Keldysh model[4] also fails to take into account the presence in the atom of a large number of excited states of the discrete spectrum near the boundary of the continuum. As a result, one can expect that the Keldysh theory should describe best of all photodetachment of electrons from negative ions, which are characterized by a short-range potential.

A comparison of the Keldysh theory and its various modifications[19,20] with numerical models of a one-dimensional atom in the multiphoton limit has already been carried out and has divulged a lack of quantitative agreement between the theory and accurate calculations in describing the photoelectron spectrum. In the present work, we restrict ourselves to a comparison of the spectrally integrated ionization probabilities. In the Keldysh theory[4] the ionization probability per unit time is given by

$$w \sim \exp\left(-\frac{2\tilde{I}}{\hbar\omega} \Gamma \right), \tag{11}$$

where

TABLE I.

$\hbar\omega$, eV	0.12	0.24	0.50	1.17	2.34
P_0 = 5 TW/cm^2	0.221	0.441	0.919	2.15	4.30
P_0 = 10 TW/cm^2	0.156	0.312	0.650	1.52	3.04

$$\Gamma = \operatorname{arcsinh} \gamma - \gamma \frac{\sqrt{1+\gamma^2}}{1+2\gamma^2},$$

and $\tilde{I}=I+e^2E^2/4m\omega^2$ is the effective ionization potential, corrected for the shift of the continuum boundary.

In the region $\gamma \ll 1$ we obtain from Eq. (11)

$$w \sim \exp\left(-\frac{4}{3}\frac{E_a}{E}(1-0.1\gamma^2) \right). \tag{12}$$

Under the conditions of our calculations, in the region $P_0 \leqslant 10^{13}$ W/cm^2 the ionization probability is proportional to the duration of the pulse. This enables us to compare calculated data with expressions (11) and (12). Noting that the parameter γ is proportional to the radiation frequency, we obtain from Eq. (12)

$$\ln w = \alpha + \beta(\hbar\omega)^2, \tag{13}$$

where α and β are parameters which depend on the radiation intensity.

In the other limiting case, $\gamma > 1$, expanding expression (11) in powers of $1/\gamma$ gives

$$w \sim \exp\left(\frac{\tilde{I}}{\hbar\omega} \right) \left(\frac{e^2E^2/4m\omega^2}{\tilde{I}} \right)^{\tilde{I}/\hbar\omega} . \tag{14}$$

As can be seen from relation (14), the power-law dependence (10) obtains only in the limit $\gamma \gg 1$, when the vibrational energy $e^2E^2/4m\omega^2$ can be neglected in comparison with the ionization potential I.

The calculated dependence of the ionization probability per pulse on the energy of a quantum $\hbar\omega$ is shown in Fig. 3 for two values of the intensity (5 TW/cm^2 and 10 TW/cm^2). Also shown are curves of the theoretical dependences (11), (12), and (14), and the corresponding values of the Keldysh parameter from Table I. As can be seen from Fig. 3, the laser field can be taken to be static in the region $\hbar\omega \leqslant 0.25$ eV, and the parabolic dependence (13) coincides with the calculated dependence up to values $\gamma \simeq 1$ (see also Ref. 5). On the other hand, the "exact" Keldysh formula and its multiphoton limit (14) also provide a good description of the probability of photoionization in the region $\gamma \geqslant 1$, integrated over the electron spectrum, if saturation of ionization in the laser pulse is not reached. In this case, in the region $\gamma \simeq 1$ both limiting cases give satisfactory agreement with the results of numerical modeling.

5. CONCLUSION

The calculations which we have carried out demonstrate the broad applicability of the theory of tunneling ionization of a quantum system with a short-range potential in the field of a light wave. The tunneling regime is easily achieved in short infrared laser pulses. In the visible and ultraviolet, as the radiation intensity increases to values corresponding to the condition $\gamma<1$, one commonly observes over-the-barrier disintegration of the atom, or negative ion, taking place during a time of the order of half an optical cycle.[21] Note that under the conditions of our calculations it is not possible to realize the regime of over-the-barrier disintegration. At the intensities that are needed in this case, namely greater than or equal to 80 TW/cm^2, ionization takes place along the leading edge of the pulse when the energy acquired by the electron upon traversing a distance $\sim d$, does not exceed the value V_0.

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