Stabilization of negative ions in ultrastrong optical fields

E. A. Volkova and A. M. Popov

Nuclear Physics Research Institute, Moscow State University, 119899 Moscow, Russia
(Submitted 23 September 1993)
Zh. Eksp. Teor. Fiz. **105**, 592–600 (February 1994)

A numerical *ab initio* solution of the time-dependent Schrödinger equation is used to treat the process of photodetachment of electrons from negative ions in an intense laser field. It is shown that when a certain critical intensity is reached, which depends on the frequency of the laser radiation, the negative ions are observed to stabilize: the probability of photodecay decreases as the radiation intensity increases. Possible mechanisms for the effect are discussed. It is shown that the stabilization effect is related to the formation of an electron wave packet oscillating with a large amplitude and with the consequent decrease in its binding to the atomic core.

Numerical *ab initio* solution of the time-dependent Schrödinger equation for a quantum system in the field of an electromagnetic wave have become one of the basic techniques for studying elementary processes in intense electromagnetic fields in recent years (see, e.g., the review of Burnett *et al.*[1] and the referenced cited there). These calculations are numerical experiments which enable us to determine the limits of applicability of various approximations customarily used in modeling, and also to treat the physics of the processes taking place under conditions such that none of the familiar approximations is justified.

Using this approach we have studied both the processes involving transition from discrete spectral states to the continuum for steady[2] and short-lived[3,4] potentials (photoionization and photodetachment) and transitions between various states of the continuous spectrum (stimulated bremsstrahlung).[5]

The limitations of existing computers generally restrict us to one-dimensional models of the phenomenon and quite short (femtosecond) durations. However, it is precisely the experiments with femtosecond pulses that enable us to study in the purest fashion the elementary interaction events, and also to observe a number of new effects, e.g., the suppression of ionization in ultrastrong optical fields.[1]

Usually the effect of suppression of ionization is related to destructive quantum interference of the amplitudes for transition to the continuum from a set of closely spaced quantum states.[6] Another possible reason for a reduction in the rate of ionization as the electric field strength of the electromagnetic wave increases is the formation of an electron wave packet oscillating with a large amplitude in the external electric field, and consequently interacting weakly with the atomic core, which reduces the probability of photon absorption. The possibility of inhibiting ionization through destructive interference was recently confirmed experimentally[7] in experiments on ionization of the Rydberg states of Ba atoms by the third harmonic of radiation from a neodymium laser with intensity $\approx 10^{11}$ W/cm^2. The second mechanism for increasing the stability of atoms requires considerably stronger fields,[1] currently greater than those attainable experimentally. Theoretical studies of the effect,[8] performed using the Kramers–Henneberg technique, show that for a given radiation frequency there exists a certain critical radiation intensity above which the ionization rate of an atom actually decreases. Su *et al.*[9] also predicted that an atom in an ultrastrong optical field would have a longer lifetime. They solved the one-dimensional time-dependent Schrödinger equation in the field of an electromagnetic wave for a one-dimensional hydrogenlike potential $V(x) \propto (1+x^2)^{-1/2}$.

Note that when intense laser radiation acts on negative ions above a certain critical value enhancement in the stability of the system against photodissociation of electrons should also be observed.[10] Here it is essential that the short-wavelength nature of the potential due to the absence of excited bound states rules out stabilization of the negative ions as a result of destructive interference. The threshold intensities of the radiation for observation of the effect may not be as large as for ionization of atoms from the ground state, since the ionization potential of most negative ions is less than 3.0–3.5 eV.

Recent work by Grobe *et al.*[11] has considered a one-dimensional numerical model of electron photodissociation from a screened Coulomb potential, which approximately describes the interaction between an electron and a hydrogen atom in an ultrastrong light field. In Ref. 11 the time dependence of an oscillating electron wave packet was analyzed as it spreads out, and the conditions were determined for the existence of this stabilization regime.

In the present work we used the algorithm for solving the time-dependent Schrödinger equation developed by Volkova *et al.*[4] to study the time dependence of the photodissociation of an electron from a negative ion in a femtosecond laser pulse as a function of the radiation intensity and the photon energy. The effect of stabilization of negative ions in fields above the critical value is considered, and possible mechanisms for this effect are discussed. The results of the calculations are compared with the data from Ref. 11.

To analyze the phenomenon theoretically we will start with the Schrödinger equation

$$i\hbar \frac{\partial \psi}{\partial t} = -\frac{\hbar^2}{2m}\frac{\partial^2 \psi}{\partial x^2} + V(x)\psi(x,t) - dE(t)\psi(x,t), \qquad (1)$$

 1063-7761/94/030315-05$10.00

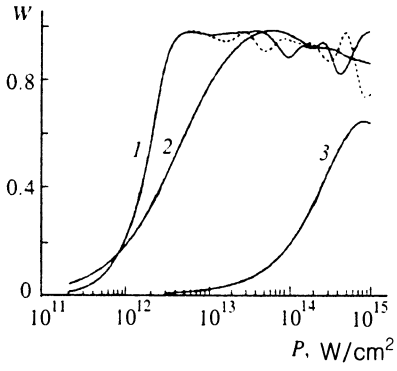

FIG. 1. Probability for photodecay of a "negative ion" as a function of the maximum radiation intensity in a pulse of 10 fs for different photon energies $\hbar\omega$, in eV: *1*) 1.17; *2*) 2.34; *3*) 5.0.

where $V(x)$ is a potential describing the interaction between the electron and the rest of the atom, $E(t)$ is the strength of the electric field of the electromagnetic wave, and d_0 is the dipole moment of the system.

We have specified $V(x)$ to have the form of a square potential well of width $l=4$ Å and depth $V_0=2.23$ eV. In such a well there exists a single bound stationary state with ionization potential $I\approx 1.9$ eV. These parameters are typical of the potentials of the negative ions of most elements.

The electric field of the electromagnetic wave is written in the form

$$E(t)=E_0\exp\left[-\frac{1}{2}\left(\frac{t-t_0}{\tau}\right)^2\right]\cos(\omega t+\varphi_0), \qquad (2)$$

which corresponds to a Gaussian laser pulse of length $t_p=2\tau$ and frequency ω; the maximum intensity of the radiation occurs at time $t=t_0$; here φ_0 is the phase of the oscillations in the field at time $t=0$.

In the calculations we set $t_p=10$–20 fs. The energy $\hbar\omega$ of a laser photon and the maximum radiation intensity $P=cE_0^2/8\pi$ varied over the ranges 1.17–5.0 eV and 10^{11}–10^{15} W/cm^2 respectively. Note the condition $\omega\tau>1$ holds over the whole range of variation of $\hbar\omega$, i.e., the laser radiation was turned on and turned off adiabatically.

At the initial time $t=0$, i.e., prior to the onset of the laser pulse ($t_0\gg\tau$) it was assumed that the system was in the unique bound state and was characterized by the wave function $\phi(x)$:

$$\psi(x,t=0)=\phi(x). \qquad (3)$$

Equation (1) was solved by means of finite elements using the technique described in Ref. 4. The spatial extent of the system was ≈ 640 Å, which enabled us to study the time dependence of the system over times $\approx 3\tau$ after the maximum intensity of the laser radiation in all intensity ranges considered.

Figure 1 shows the results of calculating the probability of electron photodissociation from a "negative ion" as a function of the maximum radiation intensity over the coarse of the laser pulse for photon energies $\hbar\omega=1.17$ (1), 2.34 (2), and 5.0 eV (3), the first and second harmonics of a neodymium laser and the frequency of a KrF excimer laser. In the first case the dissociation takes place through a two-photon process; in the other cases one photon suffices to remove an electron.

In the weak-field limit perturbation theory can be used and the probability W for photodissociation of an electron by the pulse is proportional to either the first or second power of the radiation intensity, depending on the minimum number of absorbed photons needed for the transition to the continuum. Hence in the range $P\geqslant 7\cdot 10^{11}$ W/cm^2 the probability for the two-photon process turns out to be larger than the probability for one-photon photodissociation for $\hbar\omega=2.34$ eV. The increase in the probability for photodissociation as a function of the order of the number of photons in the process has been experimentally observed[12] in connection with studies of the action of radiation with wavelengths 1064 nm and 1908 nm on negative chlorine ions.

For intensities greater than a certain critical value P^*, which depends on the laser radiation wavelength ($P^*\approx 3\cdot 10^{12}$ W/cm^2 for a neodymium laser and $P^*\approx 3\cdot 10^{14}$ W/cm^2 for a KrF laser), the power-law dependence of $W(P)$ is lost and the probability for photodissociation ceases to grow as a function of the radiation intensity. When P increases further the ion begins to stabilize in the radiation field: increasing the intensity inhibits transitions to the continuum. Threshold intensities P_s for the occurrence of the stabilization effect for a negative ion in a radiation field are shown in Table I. Note that the probability W_s for photodissociation at $P=P_s$ is practically equal to unity for a neodymium laser and its second harmonic, while for a KrF laser it is less than 0.65 (see Table I).

Special note should be taken of the nonmonotonic behavior of $W(P)$ for a neodymium laser in the range $P>P_s$. Under the conditions we considered this effect is probably related to the violation of the adiabaticity condition for the change of the electric field in a laser pulse, even though the condition $\omega\tau>1$ is satisfied. To test this hypothesis we have performed calculations of the photodissociation dynamics of an electron subject to radiation from a neody-

TABLE I.

$\hbar\omega$, eV	P_s, W/cm^2	W_s	Theoretical estimates for P_s, W/cm^2	
			Eq. (4)	Eq. (5)
1.17	$5\cdot 10^{12}$	0.970	$7.2\cdot 10^{12}$	$1.6\cdot 10^{13}$
2.34	$8\cdot 10^{13}$	0.972	$1.15\cdot 10^{14}$	$9.3\cdot 10^{13}$
5.0	$8\cdot 10^{14}$	0.650	$2.4\cdot 10^{15}$	$9.0\cdot 10^{14}$

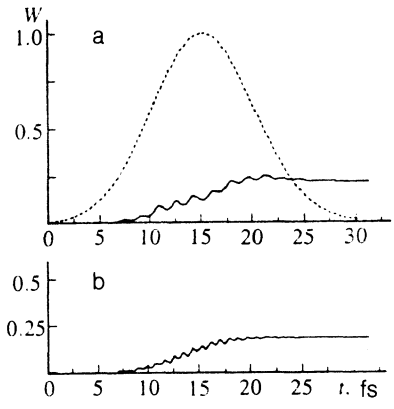

FIG. 2. Probability of observing an electron in the continuum as a function of time for $P_0=10^{12}$ W/cm^2: a) $\hbar\omega=1.17$ eV; b) $\hbar\omega=2.34$ eV. The broken trace represents the envelope of the laser pulse.

mium laser, but with the initial oscillation phase of the electric field set equal to $\varphi_0=-\pi/2$. The corresponding results are also shown in Fig. 1 (broken trace). As can be seen, a discrepancy develops only in the region $P>P_s$. Similar calculations with phases φ_0 shifted by $\pi/2$ for the cases $\hbar\omega=2.34$ and 5.0 eV, and also for a neodymium laser pulse of length 20 fs reveal that the results of calculating W do

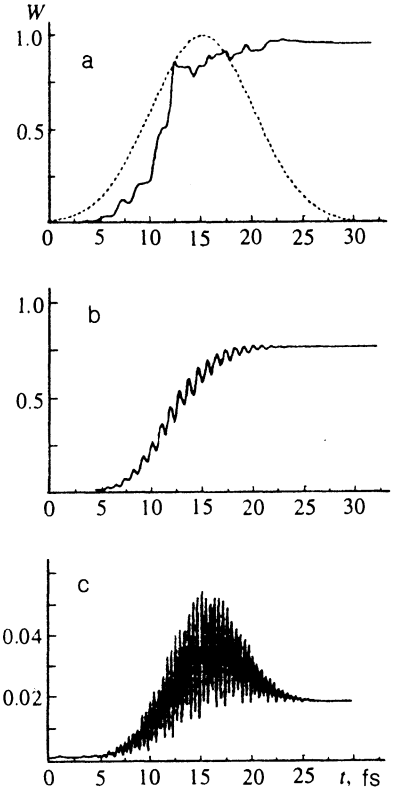

FIG. 3. Probability for observing an electron in the continuum as a function of time for $P_0=10^{13}$ W/cm^2: a) $\hbar\omega=1.17$ eV; b) $\hbar\omega=2.34$ eV; c) $\hbar\omega=5.0$ eV. The broken trace represents the envelope of the laser pulse.

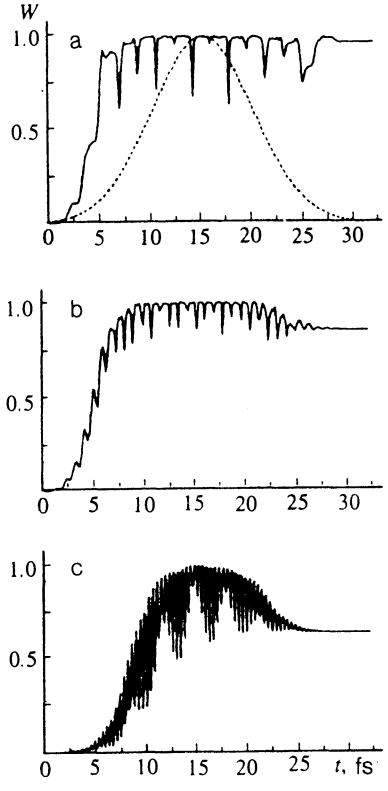

FIG. 4. Probability for observing an electron in the continuum as a function of time for $P_0=10^{15}$ W/cm^2: a) $\hbar\omega=1.17$ eV; b) $\hbar\omega=2.34$ eV; c) $\hbar\omega=5.0$ eV. The broken trace represents the envelope of the laser pulse.

not depend on the initial oscillation phase over the whole range of intensities studied.

We should say a little more in connection with possible mechanisms for suppressing the detachment process in a strong radiation field. As already noted, this system has a single bound state. Hence the ion can be stabilized by destructive interference of the transition amplitudes from different bound states to the continuum. For the same reason the observed nonmonotonic dependence of $W(P)$ cannot be due to the occurrence of a resonant transition between paired states of the discrete spectrum as a result of the dynamic Stark effect.[3]

Another possible reason for stabilization is related to the increase in the amplitude $a_e=eE_0/m\omega^2$ of the electron oscillations in the field of the electromagnetic wave to values much larger than the range l of the potential, as a result of which photon absorption becomes unlikely. Here it is necessary to point out that the absence of the long-range Coulomb "tail" of the potential, which is characteristic of the interaction between an electron and a positive ion, can reduce P_s for a short-range potential below the Coulomb value.

The condition $a_e \gg l$ enables us to estimate the threshold stabilization intensity:

$$P_s \approx m^2 c\omega^4 l^2/8\pi e^2. \qquad (4)$$

A somewhat different expression for the stabilization

TABLE II.

P, W/cm^2	$U = 1 - W$	
	t_p = 10 fs	t_p = 20 fs
$2 \cdot 10^{11}$	0.986	0.977
$1 \cdot 10^{12}$	0.789	0.659
$5 \cdot 10^{12}$	$3.35 \cdot 10^{-2}$	$6.7 \cdot 10^{-4}$
$1 \cdot 10^{13}$	$3.77 \cdot 10^{-2}$	$2.6 \cdot 10^{-3}$
$1 \cdot 10^{14}$	0.124	$1.0 \cdot 10^{-2}$

threshold follows from Refs. 13 and 14, where it was assumed that the stabilization occurs when the electron vibrational energy $\varepsilon_e = e^2E^2/4m\omega^2$ exceeds $\hbar\omega$, i.e., when the energy needed for photodetachment of an electron is substantially increased. In this case for the stabilization threshold we have

$$P_s \approx mc\hbar\omega^3/2\pi e^2. \quad (5)$$

Estimates of the stabilization threshold according to Eqs. (4) and (5) for the conditions of our experiments are also shown in Table I. As can be seen, they agree satisfactorily with one another and with the calculated results.

A quantitative comparison between the estimated expressions and the results of the numerical calculations is not straightforward, since the calculations were performed for radiation intensities varying continuously in time. Thus, in sufficiently strong fields the transition of an electron to the continuum could occur even while the intensity was increasing. This factor is clearly visible in the plots of the probability for photodissociation of an electron as a function of time over the course of the laser pulse for different intensities and radiation frequencies (Figs. 2–4). Thus, in the region $P_0 \approx 10^{12}$ W/cm^2 photodissociation occurs only at intensities close to the maximum. For $P_0 = 10^{15}$ W/cm^2, on the other hand, the probability W of observing an electron in the continuum increases almost to unity, even in the leading edge of the laser pulse. Hence $W(t)$ is found to be a strongly oscillatory function of time with a period equal to half the period of the electromagnetic wave field. From this it follows that for the effect to be experimentally observable it is necessary to try to reduce the rise time of the radiation intensity in order to avoid photodissociation in the leading edge of the pulse. Calculations of the probability that the electron will be in the bound state ($U = 1 - W$) after a pulse of length 20 fs from the Nd laser has acted on it also show the presence of an effect, but in this range of parameter the probability of photodetachment as a function of intensity increases very little in comparison with the calculations for $t_p = 10$ fs (Table II).

It is of interest to compare these results on the dynamics of photodetachment of an electron from a short-range potential with the results of similar calculations.[11] Thus, in Ref. 11 it was noted that in an ultrastrong optical field the electron wave function can be described approximately as an oscillating free wave packet spreading in space. When the laser interaction time is long, so that the region within which the electron density is localized approaches the amplitude of the free electron oscillation in the electromagnetic wave field, the atomic potential has a major effect on the evolution of ψ and a dichotomy develops.[9] We also investigated the spreading of an initial wave packet determined by the wave function of the bound state in the well in optical fields corresponding to the stabilization regime. Comparison with the results of exact calculations (Fig. 5) shows that the effect of the atomic potential on the evolution of the wave function is slight, except for the leading edge of the pulse, where the electron cannot be regarded as free.

It is interesting to note (Fig. 5) that the suppression of photodissociation occurs when the radiation pulse is dropping and is related to the "shutoff" of the electron vibrational energy as the electromagnetic field decreases adiabatically, causing the electrons to partially return to the initial bound state. In this case the probability of observing

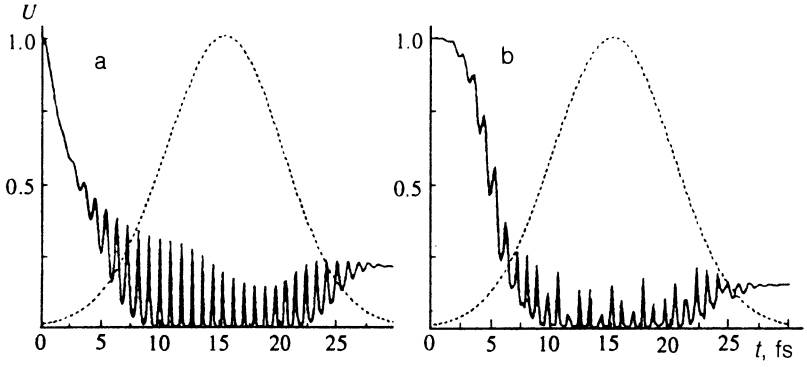

FIG. 5. Probability for observing an electron in the initial state as a function of time for $P_0 = 10^{15}$ W/cm^2, $\hbar\omega = 2.34$ eV: a) oscillations of a free wave packet; b) photodissociation of the electron from the potential well. The broken trace represents the envelope of the laser pulse.

the system in the initial state after the conclusion of the laser pulse is determined by[11]

$$U_0=\left|\int \psi^*(x)\phi_0(x)dx\right|^2, \tag{6}$$

where ϕ_0 and ψ are the wave packets of the initial and final states.

An elementary estimate of the magnitude of U_0 under the assumption that the packet broadens considerably during the time the laser pulse acts on its yields

$$U_0\approx ml^2/\hbar T, \tag{7}$$

where l is the initial width of the packet (the width of the well) and T is the pulse length. Under the conditions of our calculations ($l=4$ Å, $T=10$ fs) we find $U_0\approx 0.14$, which agrees qualitatively with the results of calculations for $\hbar\omega=1.17$ and 2.34 eV and radiation intensity $P\geqslant 3\cdot 10^{14}$ W/cm^2.

In ultrastrong optical fields the effect of the atomic potential $V(x)$ of the evolution of the oscillatory wave packet can be calculated from perturbation theory using an approach developed by Bunkin *et al.*.[15] Since under these conditions the spectral width of the initial wave packet satisfies $\delta\varepsilon\approx\hbar^2/ml^2\ll\hbar\omega$, we can assume that the perturbation $V(x)$ induces transitions between sets of closely spaced free-electron state in the electromagnetic wave field described by the Volkov functions

$$\phi_p(x)=\exp\left\{\frac{i}{\hbar}\left(p-\frac{eE_0}{\omega}\sin\omega t\right)x-\frac{i}{\hbar}\int_{-\infty}^{t}\frac{1}{2m}\times\left(p-\frac{eE_0}{\omega}\sin\omega t\right)^2 dt\right\}, \tag{8}$$

where the average momentum p_0 of the initial state ϕ_{p0} is equal to zero. The probability of the transition $\phi_{p0}\to\phi_p$ is proportional to the square of the matrix element: $W_{pp_0}\propto|\langle\phi_p|V(x)|\phi_{p0}\rangle|^2$. Assuming that the final state energy is $\varepsilon=p^2/2m\approx e^2E_0^2/4m\omega^2$, we find the following transition probability W_{pp_0} as a function of the laser intensity:

$$W_{pp_0}\approx 1/P. \tag{9}$$

From (7) and (9) we find an estimate which describes the process of stabilizing a negative ion due to wave-packet oscillations:

$$U\approx U_0(1-P^*/P), \tag{10}$$

where P^* is a characteristic intensity on the order of the stabilization threshold P_s when the ionization probability is close to unity.

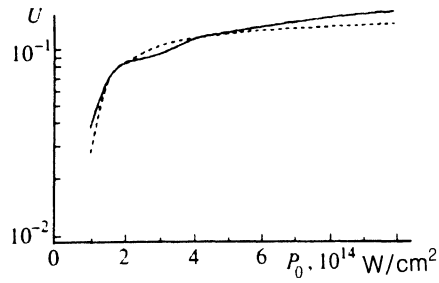

FIG. 6. Probability for observing an electron in the bound state after application of a laser pulse ($\hbar\omega=2.34$ eV) of length 10 fs as a function of the maximum intensity: solid trace) exact calculation; broken trace) estimate according to Eq. (10).

Estimates using Eq. (10) for $\hbar\omega=2.34$ eV and $P^*=P_s=8\cdot 10^{13}$ W/cm^2 are shown in Fig. 6. As can be seen, they agree well with results obtained by numerically integrating the Schrödinger equation.

In conclusion we remark that it is considerably simpler in experiments to observe the stabilization of negative ions than the analogous process of stabilizing atoms in ultrastrong light fields. The optimum laser intensities in the visible wave band are equal to 10^{14}–10^{15} W/cm^2 for pulse lengths of order 10 fs. Such laser parameters are close to those currently attainable and encourage us to believe that these experiments will be performed in the near future.

[1] K. Burnett, V. C. Reed, and P. L. Knight, J. Phys. B **26**, 561 (1993).
[2] J. Javanainen, J. H. Eberly, and Q. Su, Phys. Rev. A **36**, 3430 (1988).
[3] J. N. Bardsley, A. Szoke, and M. J. Comella, J. Phys. B **21**, 3899 (1988).
[4] E. A. Volkova, A. M. Popov, and O. B. Popovicheva, Zh. Eksp. Teor. Fiz. **102**, 496 (1992) [Sov. Phys. JETP **75**, 263 (1992)].
[5] E. A. Volkova, A. M. Popov, and O. B. Popovicheva, Fiz. Plazmy **18**, 1558 (1992) [Sov. J. Plasma Phys. **18**, 807 (1992)].
[6] M. V. Fedorov and A. M. Movsesian, J. Opt. Soc. Am. B **5**, 850 (1988); M. V. Fedorov, M. Yu. Ivanov, and A. M. Movsesian J. Phys. B **23**, 2245 (1990).
[7] R. R. Jones and P. H. Bucksbaum, Phys. Rev. Lett. **67**, 325 (1991).
[8] M. Pont and M. Gavrila, Phys. Rev. Lett. **65**, 2362 (1990).
[9] Q. Su, J. H. Eberly, and J. Javanainen, Phys. Rev. Lett. **64**, 862 (1990).
[10] P. A. Golovinskii and I. Yu. Kiyan, Usp. Fiz. Nauk **160**, 98 (1990) [Sov. Phys. Usp. **33**, 453 (1990)].
[11] R. Grobe and M. V. Fedorov, Phys. Rev. Lett. **68**, 2592 (1992); J. Phys. B **26**, 1181 (1993); Laser Phys. **2**, 265 (1993).
[12] M. D. Davidson, D. M. Schumacher, P. H. Bucksbaum *et al.*, Phys. Rev. Lett. **69**, 3459 (1992).
[13] M. Pont and R. Shakeshaft, Phys. Rev. A **44**, R4110 (1991).
[14] M. Pont, D. Proulx, and R. Shakeshaft, Phys. Rev. A **44**, 4486 (1991).
[15] F. V. Bunkin and M. V. Fedorov, Zh. Eksp. Teor. Fiz. **49**, 1215 (1965) [Sov. Phys. JETP **22**, 844 (1966)]; F. V. Bunkin, A. E. Kazakov, and M. V. Fedorov, Usp. Fiz. Nauk **107**, 559 (1972) [Sov. Phys. Usp. **15**, 416 (1973)].

Translated by David L. Book