Short-pulse detachment of H^- in the presence of a static electric field

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Quantum interference effects occurring in photodetachment of H^- in the presence of a uniform, static electric field are shown theoretically to be controllable through use of short laser pulses having characteristic times comparable to photodetached electron reflection times. In particular, calculated cross sections for single-photon detachment by two laser pulses that are delayed and phase shifted relative to one another are shown to oscillate as a function of the relative phases of the laser pulses at fixed photodetached electron energy.

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Negative ion detachment in a static electric field leads to quantum interference effects (e.g., cross section oscillations) near threshold due to the two possible paths for electron detachment along the static field direction: a direct path and a reflected path [1,2]. This interference has been observed [3]. We show here that this interference may be controlled using laser pulses with characteristic times comparable to detached electron reflection times.

Many recent works have shown that phase interference effects between different quantum mechanical pathways may be used to control the outcome of atomic and molecular processes [4,5]. Typically, such control is achieved either through interference between single and multiphoton processes [4] or else by means of time delays in resonant two-photon processes [5]. In this work, control of interference is demonstrated for a single photon process using short laser pulses.

Our calculations employ an analytical solution [6] of the time-dependent, three-dimensional Schrödinger equation (for a detached electron moving in both a laser field and a static electric field) that has been suitably generalized to treat the temporal development of the laser pulse. For a static electric field, $\mathbf{E}_S = E_S\hat{\mathbf{k}}$, and a laser pulse of frequency ω defined by the vector potential $\mathbf{A}_L = A_L(\omega,t)\hat{\mathbf{k}}$, this solution (in momentum space) is

$$\Psi_{k_xk_y\epsilon_z}(\mathbf{p},t) = (2\pi E_S)^{-1/2}\delta(p_x-k_x)\delta(p_y-k_y)e^{-i(\epsilon_\perp+\epsilon_z)t-i\phi(t)}\exp\left\{i\frac{p_z^3}{6E_S} - i\left[\frac{\epsilon_z}{E_S}+\frac{1}{c}\int^t A_L(t')dt'\right]p_z\right\}, \quad (1)$$

where

$$\phi(t) \equiv \frac{1}{2c^2}\int^t A_L^2(t')dt' + \frac{E_S}{c}\int^t dt'\int^{t'} A_L(t'')dt'' . \quad (2)$$

The subscripts k_x, k_y, and ϵ_z indicate that the electron's momentum perpendicular to the z axis and its energy along the z axis are conserved; $\epsilon_\perp \equiv \frac{1}{2}\left[k_x^2+k_y^2\right]$.

Because the H^- ion has served as a prototype system for detachment processes in the presence of a static electric field [7], we choose our initial-state wave function to be the Ohmura and Ohmura wave function for H^- [8], $\Psi_i(\mathbf{p},t) = B(2\pi)^{-1/2}\left[\frac{1}{2}p^2-\epsilon_i\right]^{-1}e^{-i\epsilon_i t}$, where $B = 0.315\,52$ a.u. and $\epsilon_i = -0.027\,751$ a.u.

The S-matrix element for the transition from this initial state to the final state in Eq. (1) is

$$S_{k_xk_y\epsilon_z} = -i\int_{-\infty}^{+\infty}\langle\Psi_{k_xk_y\epsilon_z}|H_I|\Psi_i\rangle dt , \quad (3)$$

where H_I comprises the usual terms involving $\mathbf{A}_L$ and E_S [6]. As in Ref. [6], H_I may be rewritten as $\epsilon_i - \frac{1}{2}p^2$. Carrying out the momentum space integrations in Eq. (3), we obtain

$$S_{k_xk_y\epsilon_z}(t) = iB(4/E_S)^{1/6}\int_{-\infty}^{t}\mathrm{Ai}\left[-(2E_S)^{1/3}\left(\frac{\epsilon_z}{E_S}+\frac{1}{c}\int^{t'}A_L(t'')dt''\right)\right]\exp i\left[(\epsilon_\perp+\epsilon_z-\epsilon_i)t'+\phi(t')\right]dt' , \quad (4)$$

where Ai is an Airy function. The S-matrix element in Eq. (3) is the limit of Eq. (4) as $t\to+\infty$; it is convenient to define a time-dependent S-matrix element in order to examine wave-packet behavior while the laser pulse is on.

The photodetachment cross section is given by

$$\sigma = \left(2^{9/2}\pi^{3/2}\alpha\omega/cE_0^2\right) \times\int_0^\infty d\epsilon_\perp\int_{-\infty}^{+\infty}d\epsilon_z|S_{\epsilon_\perp\epsilon_z}(t\to+\infty)|^2, \quad (5)$$

where we have replaced dk_xdk_y by $2\pi d\epsilon_\perp$ and where the factor in parentheses is the inverse of the photon flux for the pulse. For the laser field strength we employ ($E_0 = 10^5$ V/cm), the S matrix in Eq. (4) is essentially nonzero only for one-photon detachment, $\epsilon_\perp+\epsilon_z \approx \epsilon_i+\omega$.

The laser pulse shapes considered in this work are shown in Fig. 1. Figure 1(a) shows a Gaussian-shaped pulse with half-width $2/\alpha$. The photodetachment cross section for H^- calculated for laser pulse half-widths of

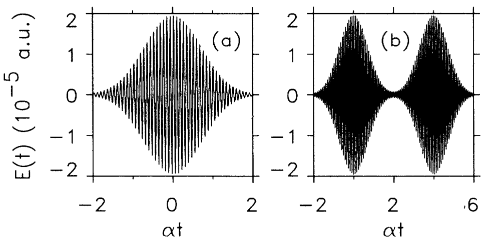

FIG. 1. (a) Gaussian-shaped single laser pulse defined by $E(t){=}E_0\exp(-\alpha^2t^2)\sin\omega t$. (b) Double laser pulse defined by $E(t){=}E_0\exp(-\alpha^2t^2)\sin\omega t + E_0\exp[-\alpha^2(t-\tau)^2]\sin(\omega t+\beta)$, where τ and β are the time delay and phase, respectively, of the second pulse relative to the first. The pulse shown has $\tau=4/\alpha$ and $\beta=0$.

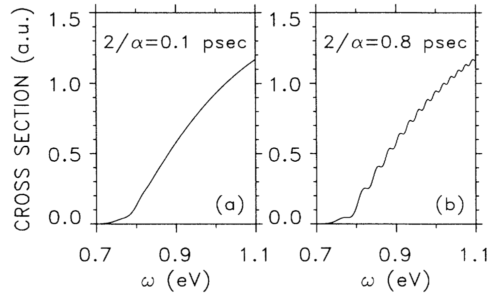

FIG. 2. Photodetachment cross section for H^- in a uniform static electric field ($E_S = 1.64\times10^5$ V/cm directed along the z axis) as produced by a single laser pulse [cf. Fig. 1(a)] linearly polarized along the z axis. The laser pulse half-width $2/\alpha$ is 0.1 psec in (a) and 0.8 psec in (b).

0.1 and 0.8 psec are shown in Figs. 2(a) and 2(b), respectively. The cross section for the longer pulse exhibits oscillations that may be described by a modulation factor nearly identical to that for an infinitely long laser pulse [9]. The shorter laser pulse, however, gives a detachment cross section which does not exhibit such oscillations. One interpretation of this result is that the shorter laser pulse is not sufficiently monochromatic; hence, interference effects at each frequency get smoothed when one averages over the frequencies effectively contained in the pulse. Another interpretation, however, based upon a time-dependent wave-packet analysis of the detached electron probability indicates the possibility for interesting new experimental measurements.

The wave-packet amplitude for the detached electron is

$$\Psi(p_x,p_y,z,t) \equiv \int_{-\infty}^{+\infty} dk_x \int_{-\infty}^{\infty} dk_y \int_{-\infty}^{\infty} d\epsilon_z \Psi_{k_xk_y\epsilon_z}(p_x,p_y,z,t) S_{k_xk_y\epsilon_z}(t)\,, \tag{6}$$

where $\Psi_{k_xk_y\epsilon_z}(p_x,p_y,z,t)$ is the z-component Fourier transform of Eq. (1) and $S(t)$ is defined in Eq. (4). For the case $p_x=p_y=0$, we plot $|\Psi(0,0,z,t)|^2$ as a function of z and t in Fig. 3 for a laser pulse [cf. Fig. 1(a)] having a half-width of $2/\alpha = 0.1$ psec. To relate the time development of the detached electron wave packet to the passage of the laser pulse over the H^- ion, we measure time in units of $1/\alpha$.

Comparing Fig. 1(a) with Figs. 3(a)–3(d), we see that when the center of the laser pulse passes over the H^- ion at $t=0$ [cf. Fig. 3(a)], the detached electron wave packet is bifurcating, with half the amplitude heading toward negative values of z and half heading toward positive values of z. The latter group, however, is trapped by the Stark potential, while the former group escapes [cf. Fig. 3(b)]. Notice, however, that for $t=2.0/\alpha$ [cf. Fig. 3(b)], the 0.1-psec half-width laser pulse has already passed over the H^- ion [cf. Fig. 1(a)].

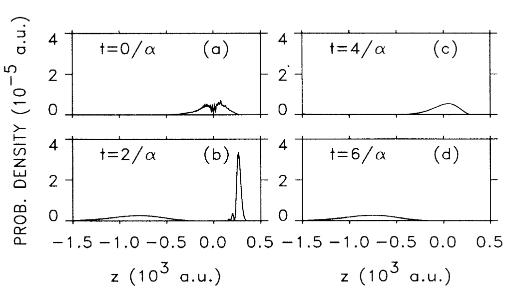

FIG. 3. Wave-packet probability $|\Psi(p_x,p_y,z,t)|^2$ for $p_x=p_y=0$ for electrons detached from H^- in a static electric field ($E_S = 1.64\times10^5$ V/cm directed along the z axis) by a laser pulse of frequency $\omega = 1$ eV having the form in Fig. 1(a), where the half-width is $2/\alpha = 0.1$ psec. Frames (a)–(d) show the electronic wave-packet distribution along the z axis for times t measured in units of $1/\alpha$. The laser pulse passes over the H^- ion during $-2.0/\alpha \lesssim t \lesssim +2.0/\alpha$; for greater times, the electronic wave packet moves only under the influence of the static field.

For the laser frequency employed in our work, $\omega = 1$ eV, the electron wave packet trapped along positive values of z takes 0.1 psec to return to the vicinity of $z=0$ [cf. Fig. 3(c)]. It then proceeds along the negative z axis [cf. Fig. 3(d)] much as the leftmost wave packet does in Figs. 3(b) but delayed by the reflection time of $\approx 4.0/\alpha = 0.2$ psec.

The wave-packet interpretation of the photodetachment cross section in Fig. 2(a) for a laser pulse of half-width $2/\alpha = 0.1$ psec is that there are no interference-induced oscillations because there is no interference. The laser pulse is gone by the time the reflected electron wave packet returns to $z=0$. Our calculations show that interference occurs whenever the pulse is long enough to still be producing detached electron probability amplitude at the time the reflected electron probability amplitude (produced by the beginning of the pulse) returns to the origin. Thus the cross section oscillations in Fig. 2(b) occur due to interference between the newly produced electron probability amplitude at the origin and the reflected probability amplitude produced at an earlier time. Since the reflection time and relative phases depend on the photon frequency ω, one observes the cross section

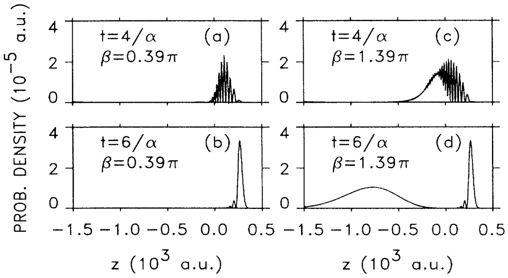

FIG. 4. Same as Fig. 3, but for the double laser pulse shown in Fig. 1(b), with each laser pulse having a half-width $2/\alpha = 0.1$ psec and frequency $\omega = 1$ eV. The time delay between pulses is $\tau = 4/\alpha = 0.2$ psec and their relative phase is β. The first pulse passes over the H^- ion during $-2.0/\alpha \leq t \leq +2.0/\alpha$ and the second pulse during $+2.0/\alpha \leq t \leq 6.0/\alpha$.

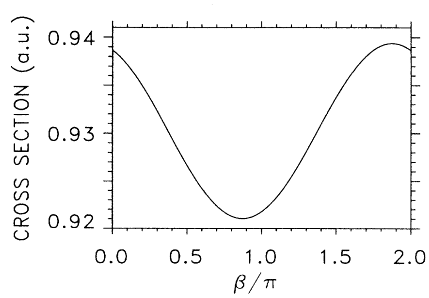

FIG. 5. Photodetachment cross section of the H^- ion in a uniform static electric field ($E_S = 1.64 \times 10^5$ V/cm directed along the z axis) by a double laser pulse of the form in Fig. 1(b) linearly polarized along the z axis. The cross section is shown for $\omega = 1$ eV as a function of the relative phase β of the two laser pulses. Each laser pulse has a half-width $2/\alpha = 0.1$ psec and they are separated by a time delay $\tau = 0.2$ psec.

oscillations as functions of ω.

This interpretation implies that one may carry out a different kind of experiment using two short (relative to the electron reflection time) laser pulses constructed so that the second pulse passes over the H^- ion at about the time the reflected electron wave packet produced by the first laser pulse returns to the vicinity of $z = 0$. Then, by varying the relative phases, one may obtain quantum interference at a fixed frequency ω as a function of the relative phase.

This is precisely the result we obtain for the double laser pulse shown in Fig. 1(b), which might be produced by splitting the Gaussian pulse shown in Fig. 1(a) and delaying and phase shifting one of the split pulses by a time τ and a relative phase β. In the calculations shown in Fig. 4 we have chosen each of the two pulses to have a half-width of $2/\alpha = 0.1$ psec, $\tau = 4/\alpha = 0.2$ psec, and $\beta = 0.39\pi$ [in Figs. 4(a) and 4(b)] or $\beta = 1.39\pi$ [in Figs. 4(c) and 4(d)]. The wave packets $|\Psi(p_x, p_y, z, t)|^2$ shown in Fig. 4 are again chosen to have $p_x = p_y = 0$. For these values of p_x and p_y, $\beta = 0.39\pi$ gives destructive interference [cf. Fig. 4(a)]; in contrast, the relative phase $\beta = 1.39\pi$ produces constructive interference [cf. Fig. 4(c)]. Both Figs. 4(a) and 4(c) show the situation at $t = 4/\alpha$, the time at which the electron probability from the first laser pulse has been reflected back to $z = 0$ [cf. Fig. 3(c)].

By the time the second laser pulse has passed over the H^- ion at $t = 6/\alpha$ [cf. Fig. 1(b)], the double laser pulse has significantly changed the amount of electron probability amplitude escaping toward negative values of z compared to the case of the single laser pulse [cf. Figs. 3(d), 4(b), and 4(d), all of which are snapshots for $t = 6.0/\alpha$]. Figure 3(d) shows the reflected electron probability from the first laser pulse escaping toward negative z; there is no second pulse. Figures 4(b) and 4(d) show that the relative phase of the second pulse relative to the first can completely destroy [Fig. 4(b)] or significantly enhance [Fig. 4(d)] the electron probability escaping toward negative z. Electron amplitude produced by the second laser pulse that proceeds directly toward negative z is interfering destructively or constructively with the reflected electron amplitude produced by the first laser pulse. The electron wave packets occurring at positive values of z for $t = 6.0/\alpha$ in Figs. 4(b) and 4(d) are produced by the second laser pulse. These wave packets will proceed for later times toward negative z, producing a distribution identical to that in Fig. 3(d) for $t = 10.0/\alpha$.

Different choices of p_x and p_y will produce interference effects such as those shown in Fig. 4 only for different values of β. The measured cross sections incorporate all of these wave packets, as indicated in Eq. (5), which integrates over all values of $\epsilon_\perp$.

The detachment cross sections resulting from the same double laser pulse used to calculate the wave packets in Fig. 4 are shown in Fig. 5 as a function of the relative phase β. By varying the relative phase β of the two laser pulses one is able to produce oscillations of the same magnitude as shown in Fig. 2(b). However, whereas those in Fig. 2(b) are produced by means of a long laser pulse (relative to electron reflection times) as a function of laser frequency, those in Fig. 5 are produced with a double laser pulse (each of short duration relative to electron reflection times) and depend on the relative phase β. Photodetachment with short laser pulses in the presence of a static electric field thus presents novel opportunities for experimental control of quantum interference effects.

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