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Control of photodetachment through a two-color low-frequency field

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We propose a photodetachment scheme for observing interference effects caused by the simultaneous action of a two-color low-frequency field with commensurate frequencies during the photodetachment event. By changing the relative phase of the low-frequency fields, considerable modifications are found to occur in the angular distributions of the photoelectrons as well as in their energy spectra. Some properties of the photoelectron angular distributions are discussed, and the extension of the energy spectra as a function of the low-frequency radiation parameters is evaluated. [S1050-2947(98)50101-1]

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The availability of coherent tunable sources has stimulated the study and the use of quantum-mechanical interference for controlling amplitude probabilities of processes involving the same initial and final states of atomic and molecular systems. In the last few years, two-color phase control of multiphoton ionization has been investigated both experimentally and theoretically. By varying the relative phase of the two fields, different pathways of excitation connecting the same initial and final states may interact, giving rise to modulation of the amplitude probability of the ionization process. Experiments involving absorption of two photons of frequency ω and one photon of frequency 2ω have been carried out by Baranova *et al.* [1], who measured modulation of the photocurrent versus the relative phase of the radiation fields, as well as polar asymmetry of the ejected photoelectrons. Asymmetries in photoelectron angular distributions have been found in two-color ionization of atomic rubidium [2]. Analogous results have been found in above-threshold ionization (ATI) experiments of krypton [3], where the atoms were submitted to the simultaneous action of a radiation field of frequency ω and its second harmonic. It is well known that in ATI experiments, carried out with a single color laser, the electron may be ejected after the atom has absorbed more photons than the minimum required to be ionized. The energy spectra of the photoelectrons show peaks that are evenly separated by a quantity $\hbar\omega$ equal to the energy of the ionizing photon, denoting that the continuum is structured by the presence of the radiation field. When a second radiation field with a commensurate frequency is added, different pathways leading the atom to the same final state may interact. For each channel of ionization characterized by the different number of photons exchanged, the result of this interaction, as shown by the experiments carried out by Schumacher *et al.* [3] gives rise to angular distributions of the photoelectrons that exhibit polar asymmetries that may be controlled by the relative phase of the two radiation fields.

A different scheme of excitation for observing interference in two-color experiments involving atomic states lying in the continuum has recently been proposed by Veniard *et al.* [4]. In this scheme the ionization process results from simultaneous exchanges (either emission or absorption) of low-frequency photons of a radiation field and the absorption of one photon of its high-order harmonics. Recently, the study of multiphoton detachment of negative ions has been, subject of both theoretical and experimental interest. In particular, experiments of two-color photodetachment of negative chlorine ions by a weak field of variable frequency $h\omega_H$ in the presence of a strong microwave [5] or infrared field [6] of frequency ω_L and amplitude E_L have been carried out in order to investigate the continuum threshold shift $\Delta = e^2E_L^2/m\omega^2$ provided by the lowest radiation field.

In the high-nonlinear regime characterized by $\Delta/h\omega_L \gg 1$, the curve showing the photodetachment cross sections as a function of ω_H exhibits ripples that were interpreted [5,7,8] in terms of interference of the direct photoelectron wave and the wave that reflects from the repulsive barrier formed by the microwave field, in analogy with the explanation given in Ref. [9], where the photodetachment in the presence of a static electric field was investigated.

In all of the above summarized schemes, the atoms are ionized by the simultaneous action of two different radiation fields having commensurate frequencies that allow the interference between different excitation pathways. It is the aim of the present Rapid Communication to propose a different scheme in which, loosely speaking, a single photon of frequency ω_H excites into the continuum a negative ion in the presence of two linearly polarized radiation fields

$$\mathbf{E}_L(t) = [E_1 \sin \omega_1 t + E_2 \sin(2\omega_1 t + \delta)]\hat{\mathbf{z}}, \tag{1}$$

having commensurate frequencies and definite relative phase δ. Hence, the high-frequency field acts as a probe testing the continuum embedded in the low-frequency fields. By varying δ, the structure of the continuum is changed and, correspondingly, the energy spectra of the ejected electrons, as well as their angular distributions, are expected to be strongly affected too. In fact, it is possible to show that the probability amplitude describing the photodetachment events caused by the absorption of a photon of frequency ω_H, in the presence of a monochromatic low-frequency radiation of frequency ω, is an even function of the average momentum $\mathbf{q}$ of the ejected electron if an odd number of low-frequency photons is exchanged, provided that the initial bound state has an even parity. It is an odd function of $\mathbf{q}$ when the number of low-frequency photons is even. Hence, the photodetachment differential cross sections turn out to be invariant under the transformation $\mathbf{q}\rightarrow-\mathbf{q}$. If a second field of double frequency 2ω is added, the active electron may be ejected through different routes. In each of them different combinations of the number of photons of frequencies ω and 2ω may be exchanged leading to the same final state. The resulting transition amplitude will be a coherent sum of partial transition amplitudes; each of them, being connected with one of the possible routes, is an odd or an even function of $\mathbf{q}$. Therefore, the angular distribution of the photoelectrons may turn out to be not invariant under the transformation $\mathbf{q}\rightarrow-\mathbf{q}$.

In our model, the electron is assumed to move in a zero-range potential

$$V(r)=\frac{2\pi}{b}\,\delta(\mathbf{r})\,\frac{\partial}{\partial r} \tag{2}$$

that supports only a bound state with energy $I_0=-(b^2/2\mathrm{m})$, which, in our calculation, will be chosen to be equal to -0.75 eV, the binding energy of H^-. The field-free ground state is taken as

$$\varphi_0(\mathbf{r})=B\left[\frac{b}{2\pi}\right]^{1/2}\frac{\exp(-br)}{r}, \tag{3}$$

where B is an empirical constant equal to $2.65^{1/2}$ (see Ref. [10]). The theory of the ionization of atoms or the photodetachment of negative ions by a weak electromagnetic field of relatively high frequency (HF) ω_H, which may be treated perturbatively in the presence of a strong low-frequency field (LF), has been developed elsewhere [11]. The extension to the case in which the photodetachment takes place in the presence of the field defined by Eq. (1) is very straightforward. Thus we quote the basic formulas giving the probability amplitude of the process in which the electron is detached in a single step after absorbing one high-frequency photon of energy $h\omega_H$ and exchanging n_1 and n_2 low-frequency photons of frequency ω_1 and ω_2, respectively (hereafter atomic units will be used),

$$T_{n_1,n_2}(\mathbf{q}_f,\delta)=\frac{\exp(in_2\delta)}{(2\pi)^2}\int_{-\pi}^{+\pi}d\alpha\int_{-\pi}^{+\pi}d\beta\; f_{n_1n_2}(\alpha,\beta)M(\mathbf{q}_f,\alpha,\beta), \tag{4}$$

with $\mathbf{q}_f$ the canonical momentum of the ejected electron, α and β the phases of the LF fields,

$$f_{n_1n_2}(\alpha,\beta)=\exp\left\{-in_1\alpha-in_2\beta+i\lambda_q(\alpha,\beta)+i\rho(\alpha,\beta)+i\,\frac{E_1E_2}{\omega_1^2-\omega_2^2}\left[\frac{\sin(\alpha+\beta)}{\omega_2}-\frac{\sin(\alpha-\beta)}{\omega_1}\right]\right\}, \tag{5}$$

$$M(\mathbf{q}_f,\alpha,\beta)=-iB\,\frac{\sqrt{b}}{\pi}\,\frac{[\mathbf{q}_f+\mathbf{K}_L(\alpha,\beta)]\cdot\hat{\mathbf{z}}E_{0H}}{\{b^2+[\mathbf{q}_f+\mathbf{K}_L(\alpha'\beta)]^2\}^2}, \tag{6}$$

$$\mathbf{K}_L(\alpha,\beta)=\left[\frac{E_1}{\omega_1}\sin\alpha+\frac{E_2}{\omega_2}\sin\beta\right]\hat{\mathbf{z}}, \tag{7}$$

the oscillating momentum imparted by the bichromatic LF field to the electron,

$$\lambda_q^L(\alpha,\beta)=\mathbf{q_f}\cdot\mathbf{K}_L, \tag{8a}$$

$$\rho(\alpha,\beta)=\frac{E_1^2}{8\omega_1^2}\sin 2\alpha+\frac{E_2^2}{8\omega_2^2}\sin 2\beta, \tag{8b}$$

$$\frac{q_f^2}{2}=\omega_H+n_1\omega_1+n_2\omega_2-|I_0|-\Delta_1-\Delta_2, \tag{9}$$

the drift energy of the electron, and Δ_1 and Δ_2 the ponderomotive potential shifts due to the field components at frequencies ω_1 and ω_2, respectively.

To arrive at Eq. (4), the weak HF field has been treated perturbatively and the final continuum states describing the ejected electron have been approximated by the nonrelativistic Volkov wave function taken, in the E gauge, as

$$\Phi_f(\mathbf{r},t)=\frac{1}{(2\pi)^{3/2}}\exp\{i[\mathbf{q}_f+\mathbf{K}_L(\omega_1t,\omega_2t+\delta)]\cdot\mathbf{r}\}\times\exp\left\{-\frac{i}{2}\int^t[\mathbf{q}_f+\mathbf{K}_L(\omega_1t',\omega_2t'+\delta)]^2dt'\right\}. \tag{10}$$

Equation (6) corresponds to the usual photodetachment matrix element describing the ejection of an electron with momentum $\mathbf{q}_f+\mathbf{K}_L(t)$, and E_{0H} is the amplitude of the HF electric field taken as

$$\mathbf{E}_H=\hat{\mathbf{z}}E_{0H}\sin\omega_Ht. \tag{11}$$

Equation (4) has been obtained for the arbitrary value of the ratio ω_2/ω_1. It is very easy to show that when ω_2 and ω_1 are incommensurate, the transition amplitude, under the transformation $\mathbf{q}_f\rightarrow-\mathbf{q}_f$, modifies as $T_{n1,n2}(\mathbf{q}_f)\rightarrow(-1)^{n1+n2+1}T(-\mathbf{q}_f)$. Therefore, the photodetachment probability results in being invariant under the inversion of $\mathbf{q}_f$, and independent of the relative phase δ. Below we concentrate our attention on the case when $\omega_2=2\omega_1$. Under this assumption, the same final electronic state may be realized by the interferences of all the possible pathways in which the numbers n_1 and n_2, associated, respectively, with the process in which photons of energy ω_1 and ω_2 are exchanged combine, giving $n_1+2n_2=N$. Hence, the probability amplitude that the electron absorbs one HF photon and exchanges the energy $Nh\omega_1$ with the bichromatic field may be expressed as the following coherent sum:

$$T_N=\sum_{n_2}T_{N-2n_2,n_2}(\mathbf{q}_N,\delta), \tag{12}$$

and the corresponding differential cross section is obtained as

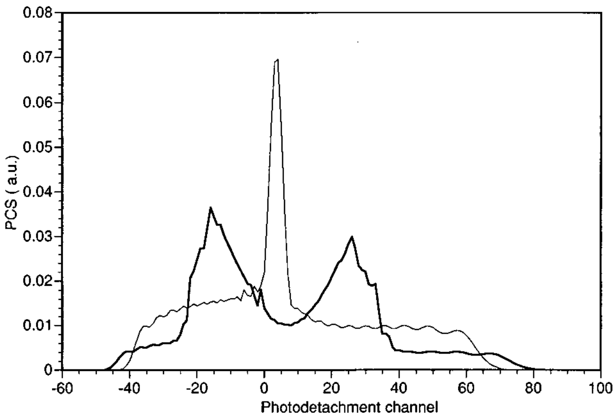

FIG. 1. Photodetachment cross sections (PCS) versus the photodetachment channel, evaluated at $\omega_H = 1.5$ eV, for two different values of the relative phase δ of the LF fields. The calculations have been carried out at the same value of the LF field amplitudes for $\omega_1 = 0.01$ eV and $\omega_2 = 2\omega_1$. The intensity of both the LF field components has been taken equal to 2×10^7 W cm^2. The lines are a guide for the eye: thin line, $\delta=\pi/2$; thick line, $\delta=0$. The amount of energy the photoelectrons exchange with the LF fields is given by $N\omega_1$.

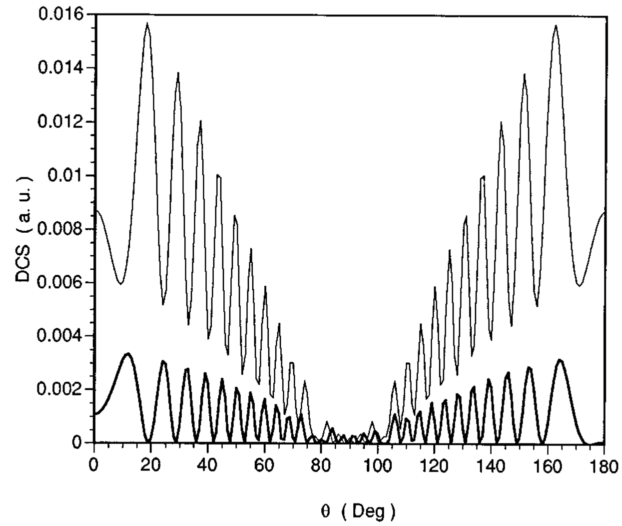

FIG. 2. Differential cross sections (DCS) of the photoelectrons emitted into the channel characterized by $N=5$ for two different values of the relative phase δ: thin line, $\delta=\pi/2$ (this distribution, when integrated over the solid angle, gives rise to the peak shown in Fig. 1); thick line, $\delta=0$. Radiation field parameters are as in Fig. 1. Note the asymmetries in the angular distribution shown by the curve evaluated at $\delta=0$.

$$\frac{d\sigma(N,\delta)}{d\Omega} = 16\pi^2 \frac{\omega_H q_N}{c} |T_N|^2, \tag{13}$$

with

$$\frac{q_N^2}{2} = N\hbar\omega_1 + \hbar\omega_H + I_0 - \Delta_1 - \Delta_2, \tag{14}$$

the drift energy of the ejected photoelectrons, and $\Delta_1+\Delta_2$ the continuum threshold shift of photodetachment. From Eq. (14) it follows that the energy spectra consist of a series of peaks evenly separated by the energy ω_1. Figure 1 shows one of these spectra obtained by integration of Eq. (13) over the solid angle, for a given value of the HF energy photon, two different values of δ, and the same value of the intensity of the LF bichromatic field components. We remark that considerable modifications occur when the value of δ is changed from 0 to $\pi/2$. For $\delta=\pi/2$ the energy spectrum shows a pronounced, narrow maximum when the photoelectron absorbs by the bichromatic field such an amount of energy $N\omega_1$ almost equal to $\Delta_1+\Delta_2$. At roughly the same energy, the energy spectra calculated for $\delta=0$ show a minimum, while two maxima located, respectively, at lower and higher energy appear. The energy separation of such maxima increases by increasing ω_H. A further distinctive feature of the energy spectra is the onset of a plateau whose extension is determined, at fixed value of ω_H, by the relative phase δ and the ponderomotive shift $\Delta_1+\Delta_2$. Calculations not reported here show that, by summing over N the cross sections shown in Fig. 1, results are obtained that are almost independent of δ. Their differences become vanishing small by increasing ω_H. In fact, independently of δ, when ω_H increases, the oscillating momentum imparted to the electron by the LF field eventually becomes a small quantity compared to the drift electron motion, and the phase effects lose their importance on the total yield. Instead, for ω_H near the photoionization field-free threshold, the total cross sections summed over N are appreciably modified by δ.

In order to better illustrate some of these features, we put Eq. (12) in the following form, valid for $\omega_2=2\omega_1$:

$$T_N = \frac{1}{2\pi}\int_{-\pi}^{+\pi} \exp\left\{+iN\alpha + i\frac{\zeta(\alpha,\delta)}{\omega_1}\right\}\widetilde{M}(q_N,\alpha,\delta)\,d\alpha, \tag{15}$$

with

$$\zeta(\alpha) = -\left(\frac{q_N^2}{2}+\Delta_1+\Delta_2\right)\alpha + \tfrac{1}{2}\int^{\alpha}[\mathbf{q}_N + \widetilde{\mathbf{K}}_L(\alpha',\delta)]^2 d\alpha', \tag{16a}$$

$$\widetilde{M}(q_N,\alpha,\delta) = M(q_N,\alpha,2\alpha+\delta), \tag{16b}$$

$$\widetilde{\mathbf{K}}_L(\alpha,\delta) = \mathbf{K}_L(\alpha,2\alpha+\delta). \tag{16c}$$

The main contribution to the integral comes from the points of stationary phase satisfying

$$\tfrac{1}{2}[\mathbf{q}_N + \widetilde{\mathbf{K}}_L(\alpha_s,\delta)]^2 = \omega_H - |I_0|, \tag{17a}$$

from which it follows that

$$q_N \cos\vartheta + \widetilde{K}_L(\alpha\delta) = \pm\sqrt{2[(\omega_H - I_0)\cos^2\vartheta - (N\omega_1-\Delta)\sin^2\vartheta]}, \tag{17b}$$

where ϑ denotes the angle between $\mathbf{q}_N$ and $\mathbf{z}$, and the signs on the right-hand side of Eq. (17a) are respectively associated with the values the projection of the kinetic momentum of the photoelectron along the z axis takes at $\omega_1 t_s = \alpha_s$. For the values of ω_H taken under consideration in the present paper, $\omega_H > I_0$, α_s may become real or complex, the integral

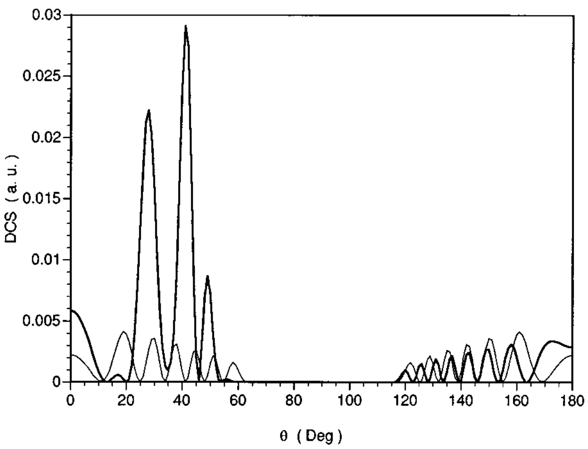

FIG. 3. As in Fig. 2 for $N=23$. For this channel the highest value of the total cross sections occurs at $\delta=0$. The extension of the angular interval in which the differential cross sections practically vanish is independent of δ.

of Eq. (15) becoming vanishing small in the latter case. This certainly happens when the square root [Eq. (17b)] becomes imaginary, i.e., when the values of ϑ fall into the angular interval defined by

$$\tan^2 \vartheta > \frac{\omega_H - |I_0|}{N\omega_1 - \Delta_1 - \Delta_2}. \quad (18)$$

Therefore, in this interval, whose extension turns out to be independent of the relative phase δ, the differential photodetachment cross sections are expected to be very small. These properties are made evident in Figs. 2 and 3, which show differential cross sections obtained by numerical evaluation of Eq. (15) for δ equal to 0 and $\pi/2$. It is to be noted that the angular distribution of the photoelectron, as well as the total cross sections resulting from angular integration of Eq. (13), are strongly affected by the variation of δ. Moreover, for $\delta=0$ the angular distributions result in being asymmetric, whereas at $\delta=\pi/2$ they are invariant under the transformation $\mathbf{q}_N \to -\mathbf{q}_N$. By Eq. (17) it is also possible to estimate the cutoff energy of the plateau that is established in the energy spectra. For real α_s, the maximum value of q_N is obtained when the canonical momentum is antiparallel to $\mathbf{K}_L$. Its modulus is very easily found to be

$$q_N = \sqrt{2(\omega_H - |I_0|)} + K_{LM}, \quad (19)$$

where K_{LM} is the amplitude of the quivering momentum imparted to the electron by the LF fields. Values of q greater than the one given by the above relation are obtained only for complex α, when the photodetachment amplitude probability becomes very small. Substitution of q_N in Eq. (14) allows us to determine the values of N beyond which the photodetachment cross sections fall by orders of magnitude. Moreover, provided that $(2\omega_H - |I_0|)^{1/2} > K_{LM}$, by replacing K_L with $-K_L$ in Eq. (18) and substituting the resulting value of q_N in Eq. (14), a value of N is found that roughly determines the lower limit of the energy spectrum. Hence, the extension of the photoelectron energy spectrum beyond which the values of the cross section fall by orders of magnitude is approximately estimated to be (in units of ω_1)

$$\Delta N \simeq \frac{2}{\omega_1} \sqrt{2(\omega_H - |I_0|)} K_{LM}. \quad (20)$$

For the cross sections shown in Fig. 1, the lower and upper values of N estimated by means of Eq. (14) become, respectively, $N=-41$ and 68 for $\delta=\pi/2$ and $N=-46$ and 79 for $\delta=0$, in very good agreement with the numerical evaluation carried out by substituting Eq. (15) in Eq. (13) and by successive integration over the solid angle.

In conclusion, we have proposed a photodetachment scheme for observing interference effects induced by the combined action of a low-frequency radiation field and its second harmonic. The extension of the energy range of the photoelectron energy spectra, as well as some properties concerning the angular distribution of the electrons, are found to be strongly affected by the relative phases of the fields.

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