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Field-assisted photodetachment process to observe the ponderomotive shift

S. Bivona, R. Burlon, G. Ferrante, and C. Leone
Dipartimento di Energetica ed Applicazioni di Fisica, Viale delle Scienze, 90128 Palermo, Italy
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Two-color photodetachment of a negative ion in the presence of a constant magnetic field is proposed as a process in which to observe the ponderomotive shift. Appropriate calculations are reported of the process under consideration based on a simple one-electron model and with the basic physical parameters well within the experimental state of the art.

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The average kinetic energy of an electron in an electromagnetic field is the sum of the average translational kinetic energy and of the so-called ponderomotive energy associated with the oscillatory motion. It is a familiar notion in the physics of charged particles interacting with electromagnetic (e.m.) fields. Accordingly, to force an electron to escape from an atom or a negative ion it is necessary to provide it with an energy greater than the binding energy by an amount equaling the ponderomotive energy, which thus may be regarded as a shift in the escape threshold. For instance, in recent years many ionization experiments have been carried out in the domain of high field intensities, showing that different groups of electrons may be ejected in the continuum, absorbing different numbers of photons. In several of such experiments the ponderomotive shift accounts for the disappearance of the low-energy peaks in the electron-energy spectrum [1]. The ponderomotive shift, however, is generally not observed in experiments in which long duration laser pulses are used, because the ponderomotive energy is converted into translational kinetic energy when the electron leaves the interaction volume. There is instead clear evidence of the ponderomotive shift in experiments with picosecond-long pulses [2].

To directly observe the ponderomotive shift, photodetachment experiments have been investigated taking the chlorine negative ion as a target and using two radiation fields. One field is of relatively high frequency ω_1, so as to strip the excess electron to the ion. In what follows the corresponding photons will be referred to as uv photons. The other field, considerably more intense, of frequency ω_2 lying in the infrared [3], is designed to determine the shift of the field-free threshold of the process. The expected shift is equal to the ponderomotive one $\Delta_2=e^2E^2/4m\omega^2$ with e and m, respectively, the absolute value of the electron charge and mass. Experiments have shown that a threshold shift is observed which is about $\frac{1}{3}$ of the value of the ponderomotive one. The results obtained with the infrared field are explained as follows [4]. The main contribution to the photodetachment rate comes from the channel for which no exchange of infrared photons occurs. This channel opens at the uv photon energy equal to the field-free photodetachment threshold $-I_0$ increased by the ponderomotive shift ($\omega_1=-I_0+\Delta_2$). The fact that the actual observed shift is smaller than Δ is then traced back to the space-time inhomogeneities of the infrared field [3,5,6].

Further ponderomotive shift measurements have been performed by Smith and co-workers [7,8]. In one experiment of two-photon detachment of H^-, a shift of about 30% of the expected value was measured [7], while in a second one [8] it amounted to 45%, with the difference in the results traced back to the uncertainties on the laser intensity. Very recently, Davidson *et al.* [6] have repeated the experiment of Trainham *et al.* [3], succeeding in separating the contributions from the different detachment channels. They report agreement with the prediction of a complete ponderomotive shift.

The above discussion shows that the direct measurement of the shift in the continuum induced by a laser field is a topic of vivid interest clearly deserving further elucidation. In this context, it is the scope of this work to describe a detachment process, allowing one to measure with sufficient accuracy the shift in the continuum. It is, in fact, well known that the photodetachment cross sections in the presence of a static magnetic field near threshold exhibit oscillations (resonances). As will be shown below, the presence of a low-frequency laser field shifts the occurrence of such resonances to values of the energy equaling the uv photon energy increased by the ponderomotive shift. The theoretical treatment is based on the formalism of the S matrix, and its main features are briefly outlined below.

Assuming a model for the negative ion as a one-electron system in which the electron moves in a static potential $V(r)$, the exact S matrix for the two-color photodetachment in the presence of a static magnetic field B is [9]

$$S_{fi}=-i\int_{-\infty}^{+\infty}\langle\Psi_f(r,t)|V(r)|\Psi_i^{\dagger}(r,t)\rangle dt, \qquad (1)$$

where Ψ_i is the exact wave function of the negative ion in the presence of two radiation fields and the magnetic field, tending, for $t\rightarrow-\infty$, to the ion bound state without the external fields. Ψ_f is the wave function of the free electron in the presence of two radiation fields and the magnetic field. Taking B along z with the vector potential, $\mathbf{A}_B=-By\hat{\mathbf{x}}$; the two radiation fields along z and y, respectively, with the vector potentials $\mathbf{A}_1=(E_1c/\omega_1)\cos(\omega_1t)\hat{\mathbf{y}}$ and $\mathbf{A}_2=(E_2c/\omega_2)\cos(\omega_2t+\varphi)\hat{\mathbf{z}}$, the wave function Ψ_f is written as

$$\Psi_f(r,t)=\left(\frac{\omega_c}{\pi}\right)^{1/4}\frac{1}{2\pi[n!2^n]^{1/2}}\exp\left\{-\frac{i}{\hbar}\left[\varepsilon_n\omega_c+\frac{p_z^2}{2m}+\Delta_1+\Delta_2\right]t\right\}\exp\{-i[\rho_2\sin 2(\omega_2 t+\varphi)+\lambda_2\sin(\omega_2 t+\varphi)]\}$$
$$\times\exp\{-i[\rho_1\sin 2\omega_1 t+\lambda_1\cos\omega_1 t]\}\exp(ip_x x)\exp\left[i\left(p_z+\frac{E_2}{\omega_2}\cos\omega_2 t\right)z\right]H_n(\zeta)\exp\left[-\frac{\zeta^2}{2}\right]$$
$$\times\exp\left[iy\left(\frac{\omega_c^2E_1}{\omega_1(\omega_1^2-\omega_c^2)}+\frac{E_1}{\omega_1}\right)\cos\omega_1 t\right], \tag{2}$$

with

$$\Delta_1=\frac{E_1^2}{4(\omega_1^2-\omega_c^2)},\quad \rho_1=\frac{\Delta_1(\omega_1^2+\omega_c^2)}{2\omega_1(\omega_1^2-\omega_c^2)}, \tag{3}$$

$$\Delta_2=\frac{E_2^2}{4\omega_2^2},\quad \rho_2=\frac{\Delta_2}{2\omega_2}, \tag{3a}$$

$$\lambda_1=\frac{\omega_c E_1}{\omega_1(\omega_1^2-\omega_c^2)}p_x,\quad \lambda_2=\frac{E_2}{\omega_2^2}p_z, \tag{3b}$$

$$\zeta=\omega_c^{1/2}y-(\omega_c)^{1/2}\left[p_x+\frac{\omega_c E_1}{\omega_1^2-\omega_c^2}\sin\omega_1 t\right], \tag{3c}$$

$$\varepsilon_n=(n+\tfrac{1}{2})\omega_c, \tag{3d}$$

where $\omega_c=eB/c$ is the cyclotron frequency, E_1 and E_2 the amplitudes of the two laser electric fields, p_x and p_z constants of motion, H_n Hermite polynomials, and n the Landau quantum number.

We now assume that (a) the intensity of the magnetic field is small enough to neglect its distorting effect on the bound state of the negative ion, and (b) the intensities of the two radiation fields produce a negligible dynamic Stark shift. With assumptions (a) and (b) $\Psi_i^\dagger$ is approximated by the wave function of the bound state in the absence of external fields. Taking, further, $V(r)$ as a three-dimensional δ function, supporting only one bound state, the spatial part of the field-free bound state is [10(a)]

$$U(r)=B_0\frac{\exp\{-\sqrt{2|I_0|}r\}}{r}, \tag{4}$$

with $I_0=-0.027\,750\,9$ a.u. the binding energy. The constant $B_0=0.315\,52$ a.u. is chosen consistently with Ref. [10(a)] and leads to a normalization of the wave function of the ground state greater than unity. We note that for this model this normalization is not unique. For a detailed discussion on this point the interested reader is referred to Ref. [10(b)]. With the above assumptions the S matrix becomes

$$S_{if}=-2\pi i\sum_{n_1}\sum_{n_2}\delta\left(\varepsilon_n+\frac{p_z^2}{2}+\Delta_1+\Delta_2-I_0-n_1\omega_1-n_2\omega_2\right)J_{n_2}(-\lambda_2,-\rho_2)M_{n_1}, \tag{5}$$

with $J_n(-\lambda_2,-\rho_2)$ the generalized Bessel function [11,12], n_1 and n_2 the numbers of photons exchanged with the two radiation fields, and

$$M_{n_1}=\frac{i^n B_0\omega_c^{1/4}}{(2^n n\sqrt{\pi})^{1/2}2\pi}\int_0^{2\pi}d\theta_1\exp[in_1\theta_1+\rho_1\sin 2\theta_1+\lambda_1\cos\theta_1]\exp\left\{-\frac{1}{2}\left(\frac{p_x}{\omega_c}+\frac{E_1\sin\theta_1}{\omega_1^2-\omega_c^2}\right)^2\omega_c\right\}H_n\left[\left(\frac{p_x}{\omega_c}+\frac{E_1\sin\theta_1}{\omega_1^2-\omega_c^2}\right)\omega_c^{1/2}\right]. \tag{6}$$

Specifying further the particular case in which (a) the radiation field along y is of low intensity and high frequency (i.e., such that the photon energy ω_1 is comparable with the electron binding energy), and (b) the field along z is high intensity and low frequency (i.e., such that the photon energy ω_2 is much smaller than the binding energy), the cross section of absorption of an ω_1 photon is written as

$$\sigma_1=\sum_{n_2}\sum_{n=0}^{n_{\max}}J_{n_2}^2(-\lambda_2,-\rho_2)\sigma_{FF}, \tag{7}$$

$$\sigma_{FF}=\frac{4\pi\omega_c A^2 I_0^2}{c\,\omega_1(\omega_1+\omega_c)^2}\left[\frac{\omega_1^2+\omega_c^2}{(\omega_1-\omega_c)^2}n+\frac{1}{2}\right]\frac{1}{p_z}, \tag{8}$$

where $A=16.079$, $n_{\max}=(n_2\omega_2+\omega_1+I_0-\Delta_2)/\omega_c-\frac{1}{2}$, and

$$p_z=\sqrt{2[n_2\omega_2+\omega_1+I_0-\Delta_2-(n+\tfrac{1}{2})\omega_c]}. \tag{9}$$

The cross section σ_1 is the incoherent sum of the cross sections of different channels, each characterized by the number n_2 of photons ω_2 exchanged with the low-frequency field and by the excited Landau level n. For each channel the

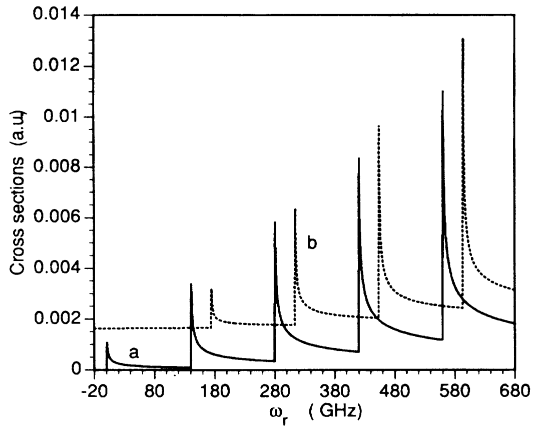

FIG. 1. Two-color photodetachment cross sections in the presence of a magnetic field as a function of the relative frequency $\omega_r = \omega_1 + I_0 - \omega_c/2$. ω_1 and ω_c are, respectively, the uv photon and the cyclotron frequencies; $-I_0$ is the electron affinity. The photon energy of the low-frequency laser is $\omega_2 = 0.1$ eV (24 178.4 GHz). The intensity of the magnetic field is $B = 5\times10^4$ G. Curve a, photodetachment without the presence of the low-frequency laser field; curve b, $I_2 = 5\times10^7$ W/cm^2.

cross section is the product of two factors: (1) $J_n(-\lambda_2, -\rho_2)$, which may be interpreted as the probability that n_2 photons of energy ω_2 are exchanged during the detachment; and (2) σ_{FF}, the detachment cross section having the same structure as that without the low-frequency field, but evaluated at p_z as given by Eq. (8). Thus, through p_z, σ_{FF} depends on the parameters of the low-frequency field as well. Keeping in mind that for $\lambda_2 \to 0$ the generalized Bessel functions are proportional to λ_2 only for odd n_2 [12], the cross sections σ_1 exhibit divergent behavior only when a channel opens with n_2 even or zero. However, such divergencies disappear if, more realistically, the line broadening of the Landau levels is taken into account.

Line broadening may be taken into account in our treatment by using a complex energy for the Landau levels:

$$\varepsilon_n = (n + \tfrac{1}{2})\omega_c + i\,\frac{\Gamma}{2}. \tag{10}$$

In such a case, in the transition probability for unit time the δ function accounting for the energy conservation is substituted by a Lorentzian, and, following known procedures [13,14], a modified cross section σ_{FF} is arrived at, the modification amounting to substituting in Eq. (8) $1/p_z$ with

$$\frac{1}{p_z} = \left[\frac{p_z^2 + \sqrt{\Gamma^2 + p_z^2}}{\Gamma^2 + p^2}\right]^{1/2}. \tag{11}$$

With this procedure, the divergencies of σ_{FF} are replaced by enhancements. We remark that the *ad hoc* introduction of the line-broadening Γ and the neglect of the radiation-atom interaction in the final wave function do not allow a detailed analysis of the behavior of the cross section as a function of energy in the regions near the Landau thresholds. On the other hand, this interesting aspect of the problem, which deserves a careful analysis, is out of the scope of the present paper and, accordingly, we will not pursue it here.

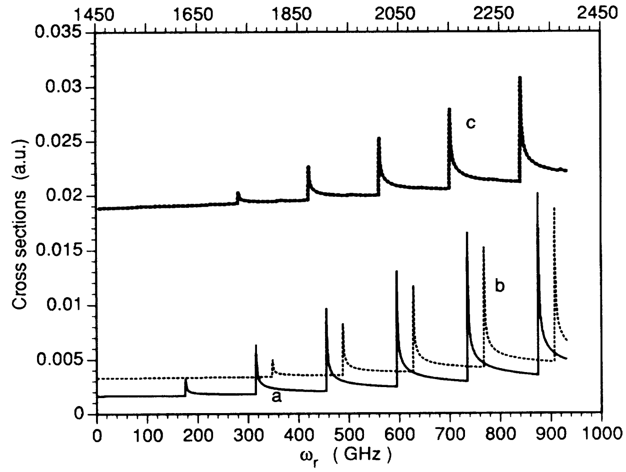

FIG. 2. Two-color photodetachment cross sections in the presence of a magnetic field as a function of the relative frequency ω_r for different values of the low-frequency field intensity I_2. The ω_r axis relative to curve c is the upper one, while that relative to the curves a and b is the lower one. For the values of the relevant parameters see caption to Fig. 1. Curve a, $I_2 = 5\times10^7$ W/cm^2; curve b, $I_2 = 10^8$ W/cm^2; curve c, $I_2 = 5\times10^8$ W/cm^2.

Before proceeding to evaluate the cross sections, Eq. (7), we discuss briefly the assumption, adopted above, of using a bare wave function to describe the negative-ion initial state; i.e., we consider the effect of the low-frequency laser field on the initial state. To this end, we take advantage of the results reported in Refs. [15] and [16], which fit our case well. According to Ref. [15] the field-dressed initial-state wave function of Eq. (1) is accurately approximated by

$$\Psi_i^\dagger = \Phi_i(r,t)\exp\left\{-i\int^t \varepsilon(\tau)d\tau\right\}. \tag{12}$$

Further, as discussed in Refs. [15,16], $\Phi_i(r,t)$ may be approximated by the wave function in the absence of the field, Eq. (4), while the second-order approximation is sufficient for $\varepsilon(t)$ [15], which is written as

$$\varepsilon(t) = I_0 - \tfrac{1}{2}\alpha_s E_2^2 \sin^2(\omega t), \tag{13}$$

with $\alpha_s = 2.65/(16|I_0|^2)$ the static dipole polarizability of the bound state [17]. By using Eq. (12) in Eq. (1), the same cross sections, Eq. (7), are arrived at with ρ_2 replaced by

$$\bar\rho_2 = (\Delta_2 - \alpha_s E_2^2/4)/2\omega \tag{14a}$$

and p_z, Eq. (9), by

$$\bar p_z = \sqrt{2\left[n_2\omega_2 + \omega_1 + I_0 - \left(\Delta_2 - \frac{a_s E_2^2}{4}\right) - (n + \tfrac{1}{2})\omega_c\right]}. \tag{14b}$$

In the following calculations, the choice of low-frequency laser parameters is such that $\alpha_s E^2/4\Delta_2$ is about 10^{-2}, so the effect of the dressing of the initial bound state by low-frequency radiation may be safely neglected.

In Fig. 1 we report the photodetachment cross section σ_1 with absorption of one uv photon vs the relative frequency $\omega_r = \omega_1 + I_0 - \omega_c/2$, and compare it with the corresponding quantity with no low-frequency field σ_{FF}. The calculation parameters are $I_2 = 10^8$ W/cm^2, $\omega_2 = 0.1$ eV (24 178.4 GHz), $B = 5 \times 10^4$ G, with I_2 the low-frequency laser intensity. The presence of the low-frequency field is found to have two effects: (a) it translates the oscillations exhibited by σ_{FF} by an amount equal to the ponderomotive shift $\Delta_2 = 347$ GHz; (b) it increases the background values of σ_1 as compared with σ_{FF}. With the chosen low-frequency field parameters, the generalized Bessel function is appreciably different from zero only for small values of n_2, and the oscillations exhibited by σ_1 correspond to uv photon energies at which the channels with $n_2 = 0$ and arbitrary Landau state number n open. According to Eq. (9), each such channel opens at the relative energy

$$\omega_r = \Delta_2 + n\omega_c . \tag{15}$$

In Fig. 1, the first oscillation occurs at $\omega_r = \Delta_2$, while the other ones are separated by ω_c. The result quoted under point (b) is explained considering that σ_1 is the incoherent sum of cross sections of different channels. In Fig. 2 we report σ_1 with the same parameters as in Fig. 1, with the only exception being for the low-frequency laser intensity, which varies from $I_2 = 5 \times 10^7$ W/cm^2 to $I_2 = 5 \times 10^8$ W/cm^2. The effect of the intensity I_2 on σ_1 is to translate the oscillations to greater values of the relative frequency ω_2 as I_2 is increased. This is in agreement with Eq. (15), according to which the values of ω_r increase linearly with Δ_2, which in its turn is proportional to I_2. Further, the curves show a highly nonlinear influence of the low-frequency field in that the values of σ_1 grow nonlinearly with the increasing of I_2. It is explained, considering that increasing I_2 increases the number of channels contributing to σ_1, that the weight of each of them, given by $J_n(-\lambda_2, -\rho_2)$, also varies nonlinearly with I_2. In this Rapid Communication we do not consider explicitly the influence of the space-time inhomogeneity of the laser beam. Obviously, the discussion given at the beginning on such an influence extends to our results as well.

In conclusion, we have reported calculations showing that using external fields with parameters well within the experimental state of the art, an independent possibility of investigating the intriguing topic of the ponderomotive shift is offered.

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