Photodetachment of H^- in the presence of a strong laser field: Effects of the laser spatial inhomogeneity

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We report on the effect of the spatial inhomogeneity of the assisting infrared radiation on the two-frequency photodetachment of H^-. We find that the curves showing the photodetachment cross section as a function of the frequency change their slope at a frequency which depends on the degree of the spatial inhomogeneity of the infrared illumination.

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INTRODUCTION

Recently, experiments [1,2] of photodetachment of negative chlorine ions by a weak ultraviolet field of variable frequency ω_H in the presence of a strong infrared field of frequency ω_L and amplitude E_L have been carried out in order to measure the threshold shift due to the presence of the assisting infrared field. The experimental results have shown that (i) the curves of the photodetachment rate versus ω_H do not exhibit a sharply defined threshold because the negative ions may simultaneously absorb one ultraviolet and one or more infrared photons; (ii) above the field-free threshold, the observed photodetachment rate is less than the field-free one and increases by increasing ω_H; (iii) the slopes of the curves showing the photodetachment cross section as a function of ω_H change very suddenly when the difference between the energy of the ultraviolet photon and the field-free energy threshold amounts to about one-third of the expected ponderomotive shift $\Delta = e^2E_L^2/(4m\omega_L^2)$.

These features have been discussed in a previous paper [3], where we calculated the photodetachment cross section under the assumption that the strong low-frequency laser field was homogeneous. In particular, we showed that the abrupt change in the slopes of the curves displaying the detachment rate as a function ω_H is due to the opening of the photodetachment channel with no exchange of infrared photons, which occurs when ω_H reaches the field-free threshold value increased by the ponderomotive shift. The difference between the expected value of ω_H and the experimental one has been related to the spatial inhomogeneity of the infrared illumination [1–3]. In fact, the negative ions that are located far from the central region of focalization, where the infrared radiation field is weaker, experience a smaller ponderomotive shift and, therefore, may be detached after absorbing only a single ultraviolet photon with an energy closer to the field-free threshold.

The aim of the present paper is to study quantitatively the effect of the spatial inhomogeneity of the low-frequency radiation on the photodetachment rate. For simplicity, we assume a one-electron model ion simulating the H^- negative ion, and use a Keldysh type approximation in the $\mathbf{E}\cdot\mathbf{r}$ gauge. In this approximation, the electron is detached through a single-step process by absorbing simultaneously the photons of both the radiation fields when it is still interacting with the residual atom [3–5]. The same physical mechanism has been recently proposed to explain the process of excess-photon detachment of negative chlorine ions [6]. In fact, the absorption of all the excess photons must occur in a very limited region, where the electron may still exchange momentum with the residual atom that, in turn, acts as a third body allowing the momentum and energy conservation.

In the Keldysh approximation [7], the ejected electron is described by a nonrelativistic Volkov plane wave, as its interaction with the residual atom is neglected. In the present paper, the distortion of the final state will be taken into account by including the phase shifts available for e-H(1s) singlet scattering [8]. This procedure has been followed by Armstrong [9] in the analysis of the H^- photodetachment and, recently, it has been reproposed by Geltmann [10] in the study of the multiphoton detachment of H^-. The results obtained by both the authors are in satisfactory agreement with the experimental data.

Finally, for our purposes, the intensity of the low-frequency laser field will be assumed to have the following Gaussian distribution

$$I_L(x,y,z) = I_M \exp(-\alpha r^2/r_0^2) .$$

I_M is the maximum intensity at the best focus of the beam, $r=(x^2+y^2)^{1/2}$ is the transverse coordinate with respect to the direction of the laser beam propagation that is assumed along the z axis, r_0 is the transverse dimension of the infrared beam, and α is a nondimensional parameter characterizing the degree of the spatial inhomogeneity of the laser. As a rule, the intensity of the high-frequency field, in any real experiment, is inhomogeneous too, and, below, the photodetachment cross sections will be obtained after averaging properly the detachment probabilities over the spatial inhomogeneities of both the radiation fields.

THEORY

Below, we outline the main steps leading to the expression of the cross section of the photodetachment in the presence of a strong radiation field. Both the radiation fields are taken to be linearly polarized and directed along z.

When the low-frequency laser field is homogeneous and taken in dipole approximation, the process may be treated in the framework of the quasienergy state. Then the amplitude probability from the bound state with absorption of one photon ω_H and exchange of n photons ω_L is obtained as [3]

$$A_{\rm qi}(n)=\langle\langle \phi_q(r,t)\exp(-in\omega_L t)|e\mathbf{E}_H\cdot\hat{\mathbf{z}}|\phi_0(r,t)\rangle\rangle \ , \qquad (1)$$

where

$$\langle\langle \cdots \rangle\rangle=(2\pi/\omega_L)\int_0^{2\pi/\omega_L} dt\langle \cdots \rangle \qquad (2)$$

and $\langle \cdots \rangle$ indicates space integration only; E_H is the amplitude of the high-frequency field; $\phi_0(r,t)$ and $\phi_q(r,t)$ are the periodic part of the quasienergy states, which tend, respectively, to the field-free ionic ground state $u_0(r)$ and to the continuum state $u_q(r)$, with kinetic energy

$$h^2q^2/2m=h\omega_H+nh\omega_L+I_0-\Delta+\delta I_0 \qquad (3)$$

when the low-frequency electric field is switched off; and δI_0 is the dynamic Stark shift of the bound state, which may be approximated, at the second order of perturbation, as

$$\delta I_0=\alpha_s E_L^2/4 \ , \qquad (4)$$

with α_s the static dipole polarizability.

The dressed bound quasienergy state may be constructed by the method of Ref. [11]. Accordingly, $\phi_0(r,t)$ may be written as

$$\phi_0(r,t)=u_0(r)\exp[-(\delta I_0/2\omega_L)\sin 2\omega_L t] \ , \qquad (5)$$

with $u_0(r)$ the spatial part of the field-free bound wave function, which, for the attractive three-dimensional δ-function potential, may be approximated as [9]:

$$u_0(r)=(2.65b/2\pi)^{1/2}\exp(-br)/r \ , \qquad (6)$$

where

$$b=(2mI_0)^{1/2} \ . \qquad (7)$$

When the interaction between the ejected electron and the atom is neglected, the continuum quasienergy state is described by the nonrelativistic Volkov wave function; accordingly, $\phi_q(r,t)$ is written as

$$\phi_q(r,t)=(2\pi)^{-3/2}\exp\{i[\mathbf{Q}_L(\omega_L t)]\cdot\mathbf{r}\} \times\exp\{i[\lambda_q(\omega_L t)+\rho(\omega_L t)]\} \ , \qquad (8)$$

$$\mathbf{Q}_L(\omega_L t)=\mathbf{q}+\mathbf{K}_L(\omega_L t) \ , \qquad (9)$$

$$\mathbf{K}_L(\omega_L t)=eE_L/(\hbar\omega_L)\hat{\mathbf{z}}\cos\omega_L t \ , \qquad (10)$$

$$\lambda_q(\omega_L t)=eE_L/(m\omega_L^2)\hat{\mathbf{z}}\cdot\mathbf{q}\sin\omega_L t \ , \qquad (11)$$

$$\rho(\omega_L t)=e^2E_L^2/(8m\omega_L^2)\sin 2\omega_L t=\rho\sin 2\omega_L t \ . \qquad (12)$$

In the field-free case, the plane-wave approximation gives a photodetachment cross section that is too small on the low-frequency side of the maximum and too large on the high-frequency side [9]. This result has been improved by using a continuum wave function that includes the phase shifts [9]. A similar procedure may be used to take into account the interaction between the ejected electron and the residual atom when a low-frequency laser field assists the photodetachment process. Accordingly, the continuum quasienergy state defined in Eq. (8) will be replaced by

$$\phi_q(r,t)=(2\pi)^{3/2}\sum_l i^{l+1}(2l+1)\exp(i\delta_l)\sin(\delta_l)[j_l(Q_L(\omega_L t)r)-i\eta_l(Q_L(\omega_L t)r)] \times\exp\{i[\lambda_q(\omega_L t)+\rho(\omega_L t)]\}P_l(\mathbf{Q}_L\cdot\mathbf{r}/Q_L r) \ , \qquad (13)$$

where j_l and η_l are spherical Bessel and Newman functions, δ_l are the phases shifts calculated at the energy $\hbar^2Q_L^2(\omega_L t)/2m$, and P_l are the Legendre polynomials.

The ansatz (13) is based on the assumption that the infrared laser field does not appreciably affect the interaction between the ejected electron and the residual atom during the photodetachment process. Moreover, the detached electron is assumed to follow the oscillations of the infrared field without any shift. A similar approximation has been used also to construct a continuum wave function of an electron in the presence of a Coulomb interaction and a low-frequency radiation field [12], and the obtained results are in good agreement with ones calculated with more refined treatments. Finally, we want to point out that, while the problem of constructing the wave function of a charged particle moving under the joint action of a potential and an electromagnetic field is still an open question, use of the ansatz provides a quite easy way for calculating the cross sections of the processes of our concern.

Proceeding in the usual way, we obtain the probability per unit time that an electron is emitted in the solid angle $d\Omega$ after absorbing one photon of frequency ω_H and exchanging n photons of frequency ω_L

$$\frac{dP}{d\Omega}=(me^2q_n/8\pi\hbar^3)|T_n(q_n,K_L)|^2E_H^2 \ , \qquad (14)$$

where

$$T_n(q_n,K_L)=\int_{-\pi}^{+\pi}d\alpha\, f_n(\alpha)M(q_n,K_L(\alpha)) \ , \qquad (15)$$

$$f_n(\alpha)=\exp[in\alpha+i\lambda_q(\alpha)-i\bar{\rho}\sin 2\alpha] \ , \qquad (16)$$

$$\bar{\rho}=\rho-\alpha_s E_L/(8\omega_L) \ , \qquad (17)$$

$$M(q_n,K_L(\alpha))=R(q_n,K_L(\alpha))\{\cos\delta_1+b[3q^2(\alpha)+b^2] \times\sin\delta_1/2q^3(\alpha)\} \ , \qquad (18)$$

$$R(q_n,K_L(\alpha))=(2.65b/\pi^3)^{1/2}\mathbf{q}(\alpha)\cdot\hat{\mathbf{z}}/[b^2+q^2(\alpha)]^2 , \tag{19}$$

$$\mathbf{q}(\alpha)=\mathbf{q}-\mathbf{K}_L(\alpha) , \tag{20}$$

$$\hbar^2q_n^2/2m=n\hbar\omega_L+\hbar\omega_H+I_0-\Delta+\delta I_0 . \tag{21}$$

The phase shifts δ_l are calculated at the energy $h^2q^2(\alpha)/2m$.

Equation (21) shows that the photodetachment channel, in which n low-frequency photons are exchanged, opens at the threshold energy given by

$$\hbar\omega_{\text{th}}(n)=-n\hbar\omega_L-I_0-\delta I_0+\Delta . \tag{22}$$

The dynamical Stark shift, for the choice of the infrared laser parameters, is found to be much less than the ponderomotive potential and, hence, it will be disregarded.

In order to account for the spatial inhomogeneity of both the kinds of radiation fields, we have to average the differential transition probability, given by Eq. (14). In fact, it depends on the distance from the laser focus through the intensities I_L and I_H of the low- and high-frequency fields, respectively. Assuming for both the fields a pulse with radius r_0 and Gaussian distribution of intensity $I_i(x,y,z)=I_{Mi}\exp(-\alpha_i r^2/r_0^2)$ $(i=L,H)$, the averaged transition probability is obtained as

$$\left\langle\frac{dP}{d\Omega}\right\rangle=(2/r_0^2)\int[dP(n,I_L,I_H)/d\Omega]r\,dr . \tag{23}$$

As the ponderomotive threshold shift changes with the distance from the laser focus, a given channel opens at different energy thresholds ω_H for different values of r. The minimum of the energy threshold occurs at $r=r_0$, and its value is

$$\hbar\omega_{\text{th}}(n)=-n\hbar\omega_L-I_0+\Delta\exp(-\alpha_L) . \tag{24}$$

Dividing Eq. (23) by the average flux of high-frequency photons and integrating over the solid angle, the averaged total cross section of photodetachment into the channel in which n photons of low frequency are exchanged is obtained as

$$\langle\sigma(n,I_L)\rangle=\int\langle d\sigma(n,I_L)/d\Omega\rangle d\Omega . \tag{25}$$

The sum over all the channels gives the averaged cross section

$$\sigma=\sum_n\langle\sigma(n,I_L)\rangle . \tag{26}$$

RESULTS AND COMMENTS

The treatment developed in the previous section is now applied to study the effects of the spatial inhomogeneity of the assisting low-frequency laser on the photodetachment process.

In our previous paper [3], we have suggested that when the nondimensional parameter $\Delta/\hbar\omega_L$ is much less than unity, a correspondence may be established between the ponderomotive shift and the photodetachment rates. If the low-frequency laser field is inhomogeneous, the ponderomotive shift becomes a function of the distance from the laser focus. Moreover, when $\Delta/\hbar\omega_L \ll 1$ at $r=0$, a perturbative regime is established in the whole region, and the photodetached electrons are ejected after absorbing few low-frequency photons. This occurs in the experiment [1,2] quoted in the Introduction, and therefore we choose to carry out calculations by assuming such laser parameters that $\Delta/\hbar\omega_L \ll 1$ in the laser focus.

In Fig. 1, we show averaged photodetachment cross sections (APCS) at different values of the parameters α_L and α_H with $r_0=100$ μm (the homogeneous case corresponds to the value $\alpha_i=0$). All the curves show a rather abrupt change in the slope when the high-frequency photon energy starts exceeding the value $\hbar\omega_0=-I_0-\Delta\exp(-\alpha_L)$, which corresponds to the field-free threshold shifted by the ponderomotive potential $\Delta\exp(-\alpha_L)$ experienced by the ions at a distance r_0 from the laser focus.

In fact, for ω_H a little larger than ω_0, the predominant channel of detachment becomes the one with $n=0$ (see Fig. 2), whose opening causes the sudden change in the slope of the curves showing the averaged cross sections. Hence, for the kind of inhomogeneity we have chosen in the present analysis, a correspondence may be established between the averaged cross section and the ponderomotive potential experienced by the negative ions in the peripheral region of illumination. In particular, assuming $\alpha_L=1$, a sizable change in the slope of the averaged photodetachment cross section occurs at an energy shifted by about one-third the field-free threshold, as in the experiment of Ref. [2].

The behavior of the cross sections shown in Fig. 1 may be explained by inspection of Fig. 3, where the photode-

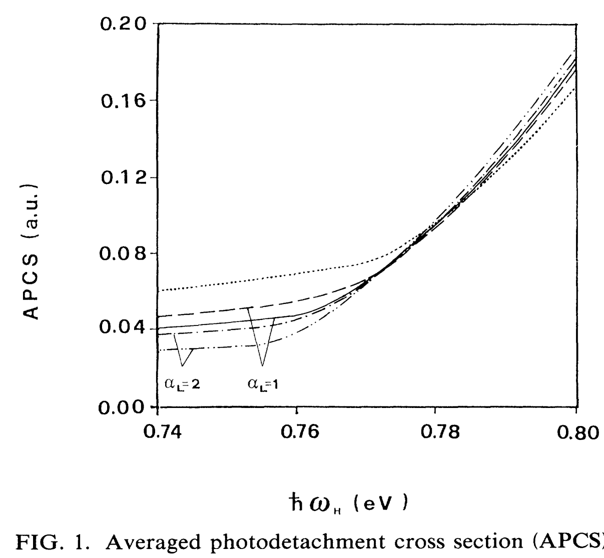

FIG. 1. Averaged photodetachment cross section (APCS) as a function of ω_H for different values of the parameter α_L and α_H. The dotted line refers to the case of a homogeneous illumination ($\alpha_L=\alpha_H=0$). Dashed and dot-dashed lines refer to the case $\alpha_H=2$. Continuous and double dotted-dashed lines refer to the case $\alpha_H=0$. The parameters of the low-frequency laser field are $I_L=5.6\times10^9$ W/cm^2 and $\hbar\omega_L=0.2$ eV. The maximum value of the ponderomotive threshold shift is $\Delta=0.02$ eV.

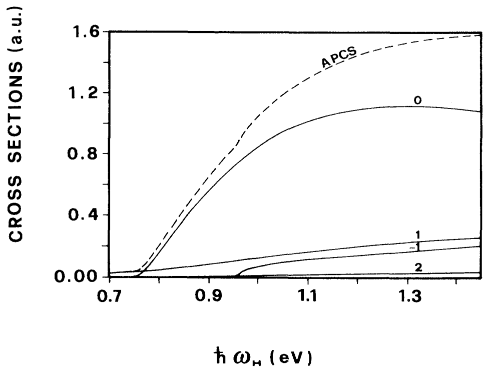

FIG. 2. Averaged photodetachment cross section (APCS) and averaged total cross sections of photodetachment, with exchange of low-frequency laser field photons as a function of ω_H for $\alpha_H=0$. The numbers on the curves denote the number of exchanged low-frequency photons. The parameters of the low-frequency laser field are $I_L=5.6\times10^9$ W/cm^2, $\hbar\omega_L=0.2$ eV, and $\alpha_L=2$.

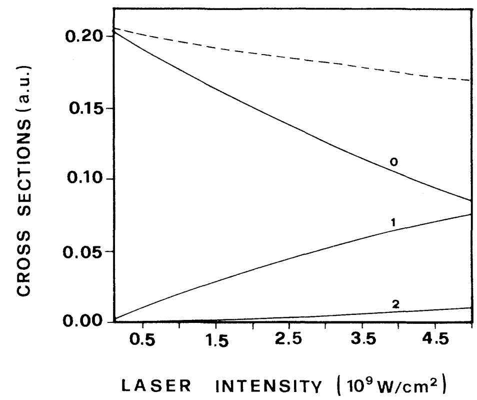

FIG. 3. Photodetachment cross section into different channels at $\hbar\omega_H=0.8$ eV vs the low-frequency laser intensity for an uniform illumination. The numbers on the curves denote the number of exchanged low-frequency photons. The dashed curve represents the sum over all the channels.

tachment cross section into different channels calculated at a fixed value of ω_H are displayed as a function of the laser intensity under the assumption of an uniform illumination. The value of ω_H has been chosen in such a way that the channels with $n>0$ are open. The cross section for $n=0$ is a decreasing function of the laser intensity, while for $n=0$ the cross sections behave in the opposite way. From this result, it follows that when ω_H is such that the channel with $n=0$ is closed, the photodetachment cross section results to be a decreasing function of the inhomogeneity parameter α_L (see Fig. 1). In fact, for increasing values of α_L, the negative ions that are located in a given region of the space experience a weaker laser intensity. Consequently, owing to the perturbative action of the infrared field, the multiphoton processes, involving an increasing number of exchanged low-frequency photons, will occur with a decreasing probability. For ω_H well above the field-free threshold, the predominant photodetachment channel is the one with $n=0$, whose opening determines heavily the behavior of the cross section summed over all the channels.

The inclusion of the spatial inhomogeneity of the high-frequency field changes the results in that the curves showing the photodetachment cross sections as a function of ω_H become slightly more smooth (see Fig. 1). In fact, in order that the channel with $n=0$ becomes the predominant one, the peripheral area from which the electron exchanging no low-frequency photons comes must increase, in order to compensate the lowering of the flux of the high-frequency radiation. Moreover, as a result, the value of ω_H at which the curves start increasing abruptly increases slightly.

In concluding, we point out that, in any real experiment, it is generally difficult to know the details of the spatial and temporal inhomogeneities of the laser beam. Therefore, while a qualitative correspondence between the measured cross section and the ponderomotive shift may be established, a precise measurement of this quantity would result to be a very difficult task.

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