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Quantum photodetachment of hydrogen negative ion in a harmonic potential subjected to static electric field

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Photodetachment of negative ions has attracted immense interest owing to its fundamental nature and practical implications with regard to technology. In this study, we explore the quantum dynamics of the photodetachment cross section of negative ion of hydrogen H^- in the perturbed one dimensional linear harmonic potential via static electric field. To this end, the quantum formula for total photodetachment cross section of the H^- ion is derived by calculating the dipole matrix element in spherical coordinates. In order to obtain the detached electron wave function, we have solved the time-independent Schrödinger wave equation for the perturbed Hamiltonian of the harmonic oscillator in momentum representation. To acquire the corresponding normalized final state detached electron wave function in momentum space, we have employed an approach analogous to the WKB (Wenzel–Kramers–Brillouin) approximation. The resulting analytical formula of total photodetachment cross section depicts interesting oscillator structure that varies considerably with incident-photon energy, oscillator potential frequency, and electric field strength as elucidated by the numerical results. The current problem having close analogy with the Stark effect in charged harmonic oscillator may have potential implications in atomic and molecular physics and quantum optics.

Keywords: photodetachment cross section, harmonic oscillator potential, Stark effect, time-independent Schrödinger equation, WKB approximation

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1. Introduction

Schrödinger wave equation has been a tremendous guide for the theoretical understanding of quantum dynamics of many quantum systems in various external potentials and fields. For instance, Schrödinger equation has been employed to understand the quantum dynamics of Stark[1] and Zeeman[2] effects. Furthermore, the electronic structures and physical properties of many-body quantum systems in condensed matter physics have been well understood from the corresponding stationary-state solution of Schrödinger equation for the energy values and corresponding wave function. However, exact analytical solution of Schrödinger equation can be found only for typical quantum systems such as electron in potential wells, hydrogen and hydrogen-like atoms and harmonic oscillator in various external fields. Photodetachment of hydrogen negative ion H^- in external fields and/or walls is yet another worth mentioning example in this regard. The photodetachment of the H^- ion in external fields and potentials is the rare example of physical problems where exact solution of Schrödinger equation can be found both semiclassically and quantically. As such, the study of photodetachment of negative ions subjected to aforementioned conditions is also a novel way for the understanding of the long-standing issue of correspondence between semiclassical and quantum mechanics.

In the earlier experiment of photodetachment of H^- in static electric field, Bryant *et al.*[3] observed interesting oscillations in the total cross section that resulted in immense experimental and theoretical development on this direction. Rau and Wong[4] explained quantum mechanically that the observed oscillations in the cross section resulted from quantum interference between the outgoing detached-electron waves emanating from the source and those reflected from the field-potential barrier. Subsequently, Du and Delos[5] studied the photodetachment of H^- in static electric field in momentum representation by solving the time-independent Schrödinger equation for the detached electron wave function. In order to further explain the origin and physical picture of the interesting scenario of the field-induced oscillations in the H^- ion photodetachment cross section, crucial development was accomplished earlier by Du and Delos[6–8] on establishing a new semi-classical approach called closed orbit theory (COT) based on reasonable approximations. According to the prediction of COT, the oscillations in cross section arise from the quantum interference between the returning detached electron waves from the walls and/or fields and the outgoing detached electron waves. Since then, the photodetachment of H^- in var-

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ious fields and/or surfaces has been studied extensively.[9–16] The experimental footprints of oscillations in the photodetachment of negative ions in electric field have been observed by the photodetachment microscopy.[17–20] Photodetachment microscopy offers a wonderful practical implementation of the photodetachment process in external fields and proved to be an innovative method for the measurement of electron affinity of atoms and direct observation of the spatial structure of detached electron wave function. To further explore the efficacy of the semiclassical approach, results of COT for some physical systems have been compared with that of quantum approach.[11,13,16,21,22] In contrast to the semiclassical approach of photodetachment which employs certain approximations, however, quantum mechanics approach provides more comprehensive results as it treats the problems at the fundamental level.

In the past, the studies on photodetachment cross section of H^- have been mostly accomplished in various fields and/or walls instead of potential wells and barriers. On the other hand, study of quantum systems in various potentials paved a way for the development of quantum mechanics and understanding of longstanding quantum phenomenon such as quantum tunneling, classical–quantum correspondence, *etc*. However, despite the significant role of potential-well problems in the understanding of many phenomena of fundamental nature and practical implications in atomic physics, nuclear physics, molecular physics, and quantum physics, photodetachment of H^- near potentials is rarely studied, with few recent exceptions.[22–26] A quantum-semiclassical comparative study of photodetachment of H^- in a one-dimensional linear harmonic potential has been recently presented by Zhao *et al.*,[22] both in position and momentum coordinates. The authors employed standard semiclassical COT and frame-transformation-theory of quantum mechanics for the study. More recently,[26] Wang and Wang investigated the photodetachment dynamics of H^- ion in a harmonic potential perturbed by a time-dependent electric field based on COT formalism.

In this paper, we extend the problem of the photodetachment H^- in harmonic oscillator potential (HOP)[22] and attempt to explore the impact of static electric field on the photodetachment cross section in HOP. In particular, we are interested to investigate how electric field strength and harmonic oscillator frequency affect the behavior of photodetachment cross section. This is among many of the rarest physical problems where exact photodetachment cross section of H^- can be obtained. The peculiarity of HOP making it a popular potential model is the fact that any general potential can be conveniently written in the form of HOP to be approximated near the equilibrium point. Besides the aforementioned points of concern, the motivation of this study also derives from the close analogy of the current problem to the Stark effect for a charged harmonic oscillator in a uniform electric field.[27] The problem has interesting implications in quantum physics and chemistry, atomic and molecular physics, and quantum optics. In addition, HOP problems have also connection regarding the measurement of the relative photoionization cross section of Rb atoms in electric fields.[28] Furthermore, the powerful predictions of the HOP model about field-dependent spacing can be effectively exploited in measuring the energy spectrum of ammonia in strong electric field.[29]

For the calculation of photodetachment cross section of H^- in HOP perturbed by the static electric field, first we will solve the time-independent Schrödinger equation for the detached electron wave function in momentum representation. Then dipole matrix element will be calculated whose modulus squared is directly proportional to quantum cross section. The impact of electric field strength, laser photon energy, and HOP frequency on the structure of photodetachment cross section will be explored with numerical results.

The layout of paper is as follows. In Section 2 we have presented the schematic geometry of the system under investigation along with its dynamical Hamiltonian. In Section 3, the outline of the basic quantum formalism is presented so as to obtain the solution of time-independent Schrödinger equation for the photo-detached electron wave function. This is followed by the calculation of total cross section. In Section 4, we present the numerical results by showing the impact of electric field strength, HOP frequency and photon energy on the cross section. In Section 5, we conclude the study with a brief summary of the findings, future prospects and implications.

We have used atomic system of units (a.u.) unless specified otherwise.

2. Schematic geometry and dynamics Hamiltonian

The proposed model of photodetachment in linear harmonic oscillator potential subjected to electric field is shown schematically in Fig. 1.[30] Let us consider the H^- ion is located in one-dimensional linear harmonic potential of the form $U(z) = \frac{1}{2}\omega^2 z^2$ confined along the z axis and is perturbed by static electric field ε which is also directed along the z axis. The electric field added a linear potential to the quadratic potential and displaced the position of the minimum of harmonic potential from O to O' as shown in Fig. 1, where the ion H^- is located. The maximum potential energy of the perturbed harmonic oscillator $\Delta E = U'(z)$ is given by $\Delta E = U'(z) - E$. The added linear potential corresponds to the external uniform field $\varepsilon = F/e$, where e is the charge of the detached electron and F is the force acting on it by the external electric field. Now consider a linearly z-polarized laser light for the photodetachment of the ion. When light falls on the ion, after absorbing

the photon energy the active electron of the ion is detached and starts moving under the influence of perturbed harmonic potential and atomic potential. The classical turning points of the perturbed harmonic oscillator are $z=\pm\sqrt{2(E+F)}/w$. The detached electron soon after leaving the vicinity of the atomic core, starts moving along $+z$ axis. However, at the classical turning point of the harmonic oscillator potential it is reflected back to the atomic core. This process is repeated and hence the detached electron executes oscillatory motion between the source and turning points of harmonic potential and hence produces oscillations in the total cross section. The detached electron may also tunnel through the potential barrier formed by the electric field and linear harmonic potential. When the electric field reaches its maximum value, the potential barrier gets strongly distorted as depicted in Fig. 1. which in turn makes the possibility of tunneling of the detached electron.

The interaction Hamiltonian of the H^- ion in harmonic oscillator potential subjected to static electric field governing the motion of the detached electron in cylindrical coordinates $(\rho z\varphi)$ is given by

$$H=\frac{1}{2}p_\rho^2+\frac{1}{2\rho}L_z^2+\frac{1}{2}p_z^2+\frac{1}{2}\omega^2z^2-Fz+V_{\rm b}(r). \tag{1}$$

Because of the cylindrical symmetry of the system, the φ motion of the detached electron can be separated from the motion in (ρ, z) plane. The symmetry of the problem renders the constancy of z component of angular momentum L_z, and we have considered the case of zero L_z for convenience. As the binding potential of the atom $V_{\rm b}(r)$ is short in range so it can be neglected when the detached electron is far from the source.

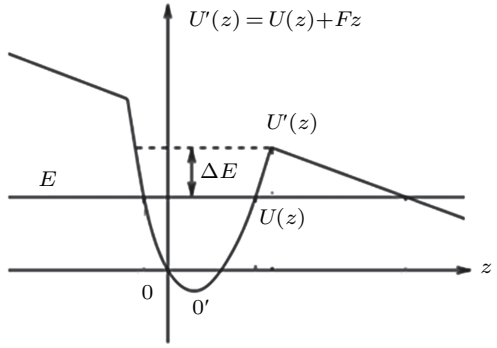


Fig. 1. Proposed photodetachment scheme in harmonic potential subjected to uniform electric field.

3. Quantum calculations of photodetachment cross section

The method we have employed in the calculation of quantum photodetachment cross section of the current system can be found in,[22] and references therein. The transition of the detached electron from the initial bound state of the H^- ion to the final continuum state is given by the quantum photodetachment cross section which is directly proportional to the square of dipole matrix element,[7]

$$\sigma=\frac{4\pi^2}{c}\int E_{\rm p}|\langle\Psi_{\rm f}^{\rm d}|D|\Psi_i^{\rm b}\rangle|^2\delta(E_{\rm f}-E)\,{\rm d}E_{\rm f}, \tag{2}$$

where D is the dipole operator, c is the speed of light with approximate value of 137 a.u.; the photon energy $E_{\rm p}$ is equal to the sum of binding energy of electron $E_{\rm b}=1/2k_{\rm b}^2$, with an approximate value of 0.754 eV, and the energy E of the photodetached electron $E=k^2/2$.

As usual, the bound-sate initial wave function of the active electron of the H^- ion in the position coordinates is given by

$$\Psi_i^{\rm b}(r)=B_0\frac{{\rm e}^{-k_{\rm b}r}}{r}. \tag{3}$$

The final-state wave function $\langle\Psi_{\rm f}^{\rm d}|$ of the photodetached electron must be energy-normalized according to the normalized condition: $\langle\Psi_{E_{\rm f}}^{\rm d}|\Psi_{E_{\rm f}'}^{\rm d}\rangle=\delta(E_{\rm f}-E_{\rm f}')$.

In order to compute the photodetachment cross section employing Eq. (2), we will first compute the dipole matrix element $\langle\Psi_{\rm f}^{\rm d}|D|\Psi_i^{\rm b}\rangle$. The initial wave function in momentum space corresponding to the wave function in Eq. (3) is given by[5]

$$\Psi_i^{\rm b}(p)=(2/\pi)^{1/2}B_0\frac{1}{\left(k_{\rm b}^2+p^2\right)}. \tag{4}$$

The final-state wave functions for the detached electron from H^- placed in one-dimensional harmonic oscillator potential in the presence of static electric field can be obtained from the solution of the Schrödinger equation in momentum space,

$$\left(\frac{1}{2}p_x^2+\frac{1}{2}p_y^2+\frac{1}{2}p_z^2+\frac{1}{2}\omega^2\frac{\partial^2}{\partial p_z^2}+{\rm i}F\frac{\partial}{\partial p_z}\right)\Psi_{\rm f}^{\rm d}(p)$$
$$=E_{\rm f}\Psi_{\rm f}^{\rm d}(p). \tag{5}$$

After solving the above equation, the final-state wave functions $\Psi_{\rm f}^{\rm d}(p)$ in momentum representation is given by

$$\Psi_{\rm f}^{\rm d}(\boldsymbol{p})=\delta(p_x-p_{x'})\delta(p_y-p_{y'})\Psi_n(p_z), \tag{6}$$

where

$$\Psi_n(p_z)=\left(\frac{1}{2^n n!}\frac{1}{\sqrt{\pi\omega}}\right)^{1/2}\exp\left[-\frac{1}{2\omega}\left(p_z-\frac{F}{\omega^2}\right)^2\right]$$
$$\times\mathbb{H}_n\left[1/\sqrt{\omega}(p_z-F/\omega^2)\right](-{\rm i})^n, \tag{7}$$

where $\mathbb{H}_n(x)$ are the Hermite polynomials.

The total final-state energy of the electron bound in harmonic oscillator potential in the presence of electric field has been lowered and is given by

$$E_{\rm f}=\frac{1}{2}p_{x'}^2+\frac{1}{2}p_{y'}^2+\left(n+\frac{1}{2}\right)\omega-\frac{F^2}{2\omega^2}. \tag{8}$$

The final-state wave function satisfies the following normalized condition:

$$\int \Psi_{\rm f}(p)\Psi'_{\rm f}(p)\,{\rm d}p = \delta(p_x - p_{x'})\delta(p_y - p_{y'})\delta_{nn'}. \quad (9)$$

By employing an approach analogues to the WKB approximation, as earlier applied by Peters and Delos,[28] the final-state wave function $\Psi_n(p_z)$ projected onto field-free states in opposite direction is given by

$$\Psi_n(p_z) = a_n\delta(p_z - p_{z'}) + b_n\delta(p_z + p_{z'}). \quad (10)$$

In order to determine the constants a_n and b_n in Eq. (10), we apply the following boundary conditions on the wave functions $\Psi_n(p_z)$

$$\Psi_n(p_z)|_{p_z=0} = [a_n\delta(p_z - p_{z'}) + b_n\delta(p_z + p_{z'})]|_{p_z=0}, \quad (11)$$

and

$$\frac{\rm d}{{\rm d}p_z}\Psi_n(p_z)\bigg|_{p_z=0} = \frac{\rm d}{{\rm d}p_z}[a_n\delta(p_z - p_{z'}) + b_n\delta(p_z + p_{z'})]|_{p_z=0}. \quad (12)$$

From the above boundary conditions, after simple calculations we can obtain

$$a_n = \left(\frac{\pi}{2.2^n n!}\sqrt{\omega/\pi}\right)^{1/2}\exp\left(-\frac{F^2}{2\omega^3}\right) \times \left[\mathbb{H}_n(F\omega^{-3/2}) - ({\rm i})p_{z'}^{-1}\omega^{1/2}\mathbb{H}'_n(F\omega^{-3/2})\right], \quad (13)$$

$$b_n = \left(\frac{\pi}{2.2^n n!}\sqrt{\omega/\pi}\right)^{1/2}\exp\left(-\frac{F^2}{2\omega^3}\right) \times \left[\mathbb{H}_n(F\omega^{-3/2}) + ({\rm i})p_{z'}^{-1}\omega^{1/2}\mathbb{H}'_n(F\omega^{-3/2})\right]. \quad (14)$$

Therefore, the final-state wave function in Eq. (6) is approximated as

$$\Psi^{\rm d}_{\rm f}(\boldsymbol{p}) = [a_n\delta(p_z - p_{z'}) + b_n\delta(p_z + p_{z'})] \times \delta(p_x - p_{x'})\delta(p_y - p_{y'}). \quad (15)$$

For z-polarized laser photon, the modulus squared of the dipole matrix element is given by

$$|\langle\Psi^{\rm d}_{\rm f}(\boldsymbol{p})|D|\Psi^{\rm b}_i(\boldsymbol{p})\rangle|^2 = |\langle\Psi^{\rm d}_{\rm f}(\boldsymbol{p})|{\rm i}\nabla_{p_z}|\Psi^{\rm b}_i(\boldsymbol{p})\rangle|^2, \quad (16)$$

where $\nabla_{p_z} = {\rm d}/{\rm d}p_z$.

Substituting the values of $\Psi^{\rm d}_{\rm f}(\boldsymbol{p})$ and $\Psi^{\rm b}_{\rm f}(\boldsymbol{p})$ in Eq. (16) and after mathematical manipulation, we obtain

$$\left|\langle\Psi^{\rm d}_{\rm f}(\boldsymbol{p})|{\rm i}\frac{\rm d}{{\rm d}p_z}|\Psi^{\rm b}_{\rm i}(\boldsymbol{p})\rangle\right|^2 = \frac{16Bo^2}{\left(k_b^2+k^2\right)^4}\frac{\omega^{1/2}}{\sqrt{\pi}2^n n!}\exp\left(-\frac{F^2}{2\omega^3}\right) \times \left[-\frac{F}{\omega}\mathbb{H}_n(F\omega^{-3/2}) + \omega^{1/2}\mathbb{H}'_n(F\omega^{-3/2})\right]^2. \quad (17)$$

After substituting the above calculated value of modulus squared of dipole matrix element into Eq. (2) and integrating, the photodetachment cross section for the z-polarized laser light in momentum representation given by

$$\sigma^z(E,F,\omega) = \sigma_0(E)\frac{3\sqrt{\pi\omega}}{(2E)^{3/2}} \times \exp\left(-F^2\omega^{-3}\right)\sum_{n=1}^{n_{\rm max}}\left(\frac{1}{2^n n!}\right) \times \left[\sqrt{\omega}\mathbb{H}'_n(F\omega^{-3/2}) - \frac{F}{w}\mathbb{H}_n(F\omega^{-3/2})\right]^2, \quad (18)$$

where the prime symbol on Hermite polynomial in the first term under summation represents its derivative with the respective variable. The smooth cross section $\sigma_0(E)$ in the field-free space is given by

$$\sigma_0(E) = \frac{16\pi^2\sqrt{2}B^2}{3c}\frac{E^{3/2}}{(E_{\rm b}+E)^3}. \quad (19)$$

Obviously, in the zero external static field case, our theoretical formula for the photodetachment cross section in Eq. (18) recovers to the one obtained earlier in Ref. [22]. The maximum number of energy levels of the electron in the harmonic oscillator potential of energy E and static field F is given by

$$n_{\rm max} = {\rm Int}\left(E/\omega + F^2/2\omega^3 - 1/2\right). \quad (20)$$

4. Numerical results and interpretation

Now we elaborate numerically the impact of electric-field strength, harmonic potential frequency, and incident photon energy on theoretical photodetachment cross section obtained earlier with formula given in Eq. (18). Figure 2 demonstrates the variation of total cross section with respect to photon energy at various fixed values of electric-field strength and at a constant oscillator frequency $\omega = 0.004$ a.u. in a particular photon energy range. It can be observed that the amplitude of cross section near threshold changes considerably with increase in electric field strength. On the other hand, the tail of cross section towards increasing photon energy end decreases gradually to a minimum value. The regular staircase shape of cross section is also disturbed with increase in electric field. Thus, electric field can control the effective value of cross section while keeping other spectral parameters fixed. In particular, the decrease in cross section amplitude with increase in field strength is controlled by the presence of electric field term in the numerator of last term in Eq. (18). The cross section is hardly affected by the field term present in the exponential term or in the argument of Hermite polynomial except at a very large value of field. The above threshold sudden decrease in amplitude of cross section is followed by the photon energy term $(E_{\rm b}+E)^3$ present in the denominator of Eq. (18). While near the threshold the cross section follows Wigner's

law where $\sigma \propto (E)^{3/2}$. The oscillations in total cross section arise after the detached electron waves are reflected from the potential barrier and interfere with the outgoing electron waves being emitted from the source.

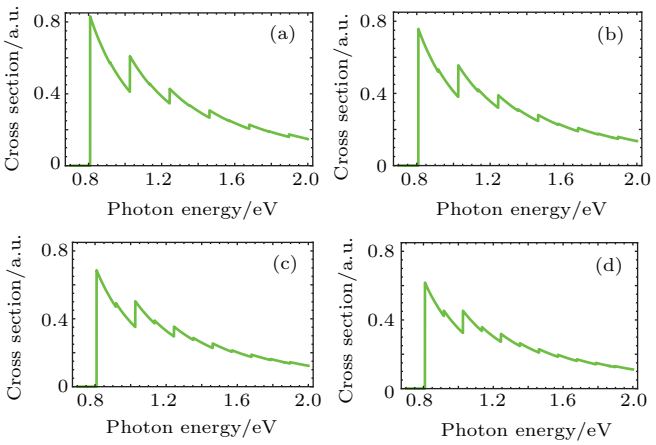


Fig. 2. Dependence of photodetachment cross section of H^- on incident photon energy at a fixed value of linear harmonic oscillator frequency $\omega = 0.004$ a.u. and at different fixed values of electric field strengths: (a) 100 kV/cm, (b) 150 kV/cm, (c) 200 kV/cm, and (d) 250 kV/cm.

In Fig. 3, we demonstrate the variation of photodetachment cross section with respect to various fixed values of harmonic oscillator frequency at constant field strength of 100 kV/cm within a specific photon energy range. The increase in the oscillator frequency besides increasing the amplitude of oscillatory cross section also increases the step size of the staircase shape of the cross section. The overall trend in change of cross section with increasing frequency is analogous to that in Fig. 2 with increasing field strength but in reverse fashion.

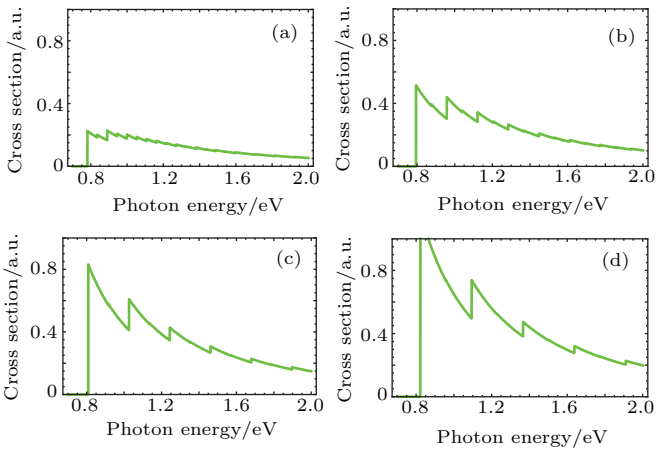


Fig. 3. Dependence of photodetachment cross section of H^- on incident photon energy at a fixed electric field strength of 100 kV/cm and at different fixed values of oscillator frequency: (a) $\omega = 0.002$ a.u., (b) $\omega = 0.003$ a.u., (c) $\omega = 0.004$ a.u., and (d) $\omega = 0.005$ a.u.

Figure 4 elucidates the variation of total cross section with respect to oscillator frequency at various fixed values of photon energy and fixed electric field strength 100 kV/cm. The cross section is endowed with a sharp but small size ripple structure, while its amplitude increases linearly with oscillator frequency at particular values of electric field strength and photon energy. However, the maximum value of cross section is shifted towards high frequency end with gradual increase in the incident-photon energy.

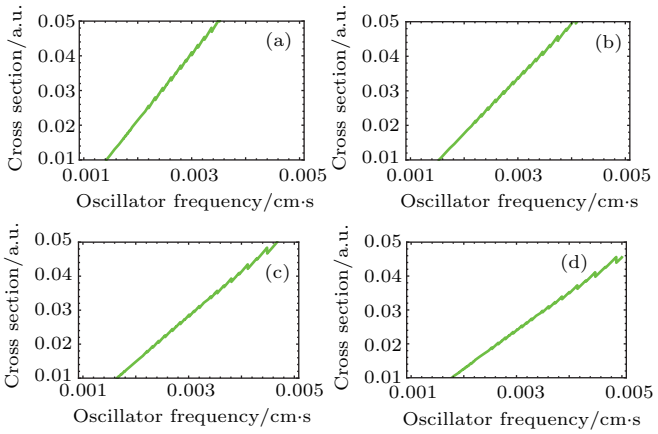


Fig. 4. Dependence of photodetachment cross section of H^- on oscillator frequency at a fixed electric field of strength 100 kV/cm and various photon energies: (a) 3.0 eV, (b) 3.25 eV, (c) 3.50 eV, and (d) 3.75 eV.

In Fig. 5, we have plotted the photodetachment cross section with respect to electric-field strength at various fixed values of incident photon energy and at a fixed oscillator frequency $\omega = 0.0005$. As can be observed in Fig. 5(a), when the incident photon energy is 1 eV, which is just above the binding energy of H^-, *i.e.*, 0.75 eV, cross section shows almost a smooth Gaussian-like shape with small width and peak amplitude. The cross section starting from a minimum zero value reaches a maximum value of 0.7 a.u. and falls again to its minimum value. In fact, as we increase the strength of electric field magnitude of cross section increases and attains maximum value at a particular value of electric field owing to dominant role of second term in square bracket of cross section given in Eq. (18). However, afterwards decrease in the cross section arises from the dominant role of first term in square bracket of cross section in Eq. (18). By further increasing photon energy at the fixed oscillator frequency, the cross section starts to display oscillations, while the peak amplitude of cross section gets maximum value at 1.5 eV, which starts to decrease again. Besides, the Gaussian shape of cross section is also disturbed after 1.5 eV. The interplay of electric field, oscillator frequency, and photon energy to modify the structure of cross section is dictated by the competitive relation of these quantities in Eq. (18). As far as the impact of photon energy on cross section is concerned, it is controlled by the term $(E_b + E)^3$ in the denominator of Eq. (19). However, the maximum value of cross section seems to originate at the particular values of the spectral parameters where their mutual harmony is optimum.

The interesting modification of cross section with oscillator frequency and field strength in the perturbed harmonic oscillator linear potential while keeping other spectral parameters fixed is observed first time in this study, which needs further investigation by closed orbit theory to conform the obtained results.

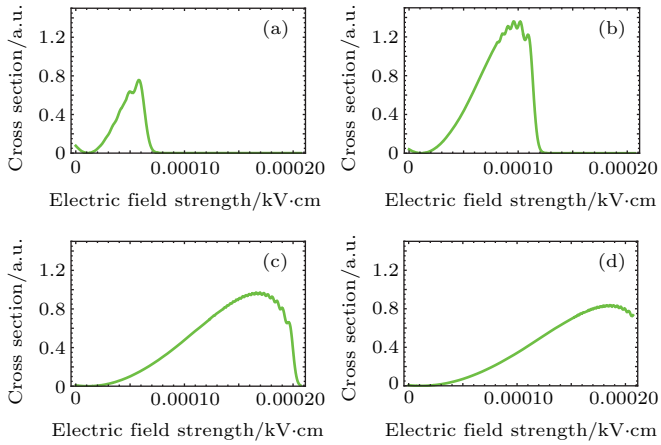


Fig. 5. Dependence of photodetachment cross section of H^- on electric field strength at a fixed value of oscillatory frequency $\omega = 0.0005$ a.u., and at various fixed values of incident photon energy: (a) 1 eV, (b) 1.5 eV, (c) 3 eV, and (d) 3.5 eV.

5. Conclusion

In conclusion, in this study we extended the problem of quantum photodetachment cross section of hydrogen negative ion confined in a one-dimensional harmonic potential to the case when it is perturbed by static electric field. The impact of photon energy, strength of electric field, and angular frequency of oscillator potential on photodetachment has been explored in detail with numerical results. In order to obtain the final-state wave function of the detached electron, we first solved the time-independent linear Schrödinger equation for the perturbed harmonic oscillator via static electric field in momentum representation. To acquire the normalized final-state wave function in momentum coordinates, we employed an approach analogous to the WKB approximation. The calculated photodetachment cross section is endowed with irregular oscillatory pattern that varies considerably with incident photon energy, the strength of static electric field and frequency of harmonic potential. A considerable modification in the amplitude and oscillatory behavior of cross section has been observed by changing the photon energy, oscillator frequency and electric field strength while keeping other spectral parameters fixed. Because of fundamental nature of HOP and its practical implications in quantum physics, quantum chemistry, atomic and molecular physics, and quantum optics, our results might be crucial for many applications in the above mentioned areas and might motivate further research on this direction. For instance, the findings might have practical implications towards understanding the complex quantum systems in the presence of potentials and electromagnetic fields with possible applications in cavity dynamics. To explore further the expected classical–quantum correspondence of the present system, the semiclassical closed orbit theory can be exploited to determine the photodetachment cross section in future.

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