Interferences in photo-detached electron spectra from a non-collinear tri-atomic anion

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The electron flux oscillations in photo-detachment of a non-collinear tri-atomic anion have been studied by taking each atom of the system as a coherent source of detached-electron wave. These electron waves traversing along three different trajectories result in a quantum interference. An analytical expression of detached-electron flux is evaluated for various detached-electron energies and for different geometrical shapes of the system. The results show that the electron flux distributions exhibit molecular shape-induced oscillatory structures. These oscillations are explained using the semi-classical closed-orbit theory; the outgoing electron waves produced from one center are propagated in the vicinity of the sources at other centers. It is also observed that in a particular case our non-collinear tri-atomic system reduces to the collinear tri-atomic system recently published.

Keywords: electron flux, collinear tri-atomic system, quantum interference

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1. Introduction

The quantum interference phenomena of microscopic objects, such as electrons and photons, etc., have played central role in the development of quantum mechanics. The grooming phenomena of quantum interference of microscopic particles have always been a subject of prime interest in quantum mechanics, not just because of its large applications, but also due to its exceptionally interesting of physics. Therefore, it has remained a focus research subject in experimental and theoretical investigations. The waves associated with microscopic particles have been thoroughly investigated in the last century. The simplest one among all the experiments describing the quantum interference phenomenon of microscopic objects is Young's double slit experiment. Young's double slit experiment explains in a simple manner the interference effects in electrons. The same interference phenomenon is observed in photo-detachment of negative ions system.

The physics of negative ions has attracted much attention in recent years. The photo-detachment of negative ions has been a focus area of research in the last few decades for structural, detached-wave function nature determination, and hence photo-detachment microscopy. Photo-detachment microscopy of negative ions has itself been established as a very sensitive technique for high accuracy measurements of the electron affinities of atoms.[1,2] Some of the results of the photo detachment of negative ions have been reported in Refs. [3]–[9]. Bryant *et al.*[3] was the first to observe a ripple-like structure in the photo-detachment cross section of H^- in the presence of a static electric field of a few hundred kV/cm above the threshold. Many theoretical studies have been presented to explain this ripple-like structure. Rau and Wong[5] used the frame transformation theory, while Du and Delos[10–12] have developed a standard closed orbit theory for the explanation of these oscillatory structures.

Afaq and Du have developed two-center model for the photo-detachment of H_2^- in free space.[13–15] Afaq *et al.*[16] have extended this idea and have presented three-center model to explain linearly organized tri-atomic anions such as $BeCl_2^-$, HCN^-, CS_2^-, and CO_2^- for z-polarized laser light. Recently, Yang and Du[17] generalized the three-center model for an arbitrary laser polarization direction and showed a strong oscillations when the laser polarization is parallel to the system axis and oscillation amplitude gradually decreases to zero as the laser polarization is changed to perpendicular to the molecular axis. Rahman *et al.*[18] extended the idea and studied photo-detachment of a homo-nuclear collinear tetra-atomic negative molecular ion.

In all of the previous researches, a particular collinear geometry of a homo-nuclear tri-atomic molecule has been considered for the detached-electron flux oscillations. To the best of our knowledge, no work has been reported about the photo-detachment of non-collinear tri-atomic negative ions. In this article, we take a non-collinear tri-atomic molecule and study the geometrical dependent electron flux oscillations first time. We shall derive an analytical formula for detached-electron flux distributions on a distant screen for different photon energies, various geometries of the system and for other parameters characterizing the tri-atomic anion. To derive the for-

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mula, we take each atom of the system as a coherent source of detached-electron waves similar to three-center model.[16] The superposition of these three waves would result in quantum interference. It will be shown that this differential cross section displays strong geometry-dependent oscillations on an observation plane. These oscillations are explained according to the closed orbit theory as: the outgoing detached-electron waves, produced from one center are propagated in the vicinity of the sources at other centers cause these oscillations. The structural information of non-collinear tri-atomic anions may be explained by analyzing these oscillations in electron flux spectra. Our derived formula for electron flux is universal, which may be applicable to any geometry of the tri-atomic molecular negative ion.

Non-collinear tri-atomic molecules like SiO^-, H_2O^-, NO_2^-, N_3^-, SO_2^-, ClO_2^-, CNS^-, NH_2^-, etc., are attractive systems due to their unique structure and potential applications. These molecules are composed of a non-collinear chain of three atoms lying in the non-collinear fashion. The knowledge of the structure and dynamics of these fascinating non-collinear tri-atomic molecules is important in quantum chemistry and molecular physics. A non-collinear chain of three hydrogen-like atoms with an extra electron (D_3^-) is assumed to be a generic model for a non-collinear tri-atomic negative ion. In this paper, the idea of quantum interference is applied to a detached-electron from the system.

This article is organized as follows: in Section 2, theoretical study is presented for the photo-detachment of a non-collinear tri-atomic negative ion, which is then applied on H_3^- anion. The results and discussion for various detached energies are presented in Section 3. Conclusions are given in Section 4. Atomic units (a.u.) will be used unless otherwise noted.

2. Theory

The non-collinear tri-atomic anion interacting with a z-polarized laser light is shown schematically in Fig. 1. The numbers 1, 2, and 3 are supposed to represent three atomic centers in the system. It is convenient to fix atoms 1 and 2 on the y axis while the atom 3 is taken variable on the z axis. O is the origin of coordinates. Let $2d$ be the distance between two adjacent atoms on y axis and x be the distance between fixed to variable atoms. The laser polarization is along z axis.

We suppose that there is only one active electron in our non-collinear tri-atomic system for the photo-detachment process. The active electron is loosely bound by a short-range spherically symmetric molecular core potential $V(r)$, where r is the distance between the active electron and origin of the system. The photo-detached electron wave function satisfies the Schrödinger equation with a source and is given by[7]

$$(E-H)\psi_{\rm D} = D\psi_{\rm I}. \tag{1}$$

In the above equation E is the detached electron energy, D is the dipole operator, $D = z$ for z-polarized light. H is the Hamiltonian that describes the behavior of our system and $\psi_{\rm I}$ is the initial wave function of the detached electron. It is also assumed that the normalized bounded initial wave function for the active electron in the system can be written as a linear combination of atomic orbital of H^--like bounded state[6] at the three centers $\psi_{\rm I} = \frac{1}{\sqrt{3}}\sum\phi_i$. Here

$$\phi_i = B\frac{\exp(-k_{\rm b}r_i)}{r_i}, \quad (i = 1,2,3),$$

are the normalized wave functions for H^- but centered at 1, 2, and 3 as shown in Fig. 1. While B is the normalization constant[6] and $k_{\rm b}$ is related to the binding energy $E_{\rm b}$ of H^- by $E_{\rm b} = k_{\rm b}^2/2$. The photo-detachment process accomplishes in two steps: in the first step, the photon energy $E_{\rm ph}$ is absorbed by an active electron and then it escapes from the negative ion as an electron wave. In the second step, the outgoing wave propagates out to a large distance. This electron wave may originate from each center of the system but with a different phase. The quantum interference of these outgoing waves at a large distance would result in oscillations in the electron flux spectra.

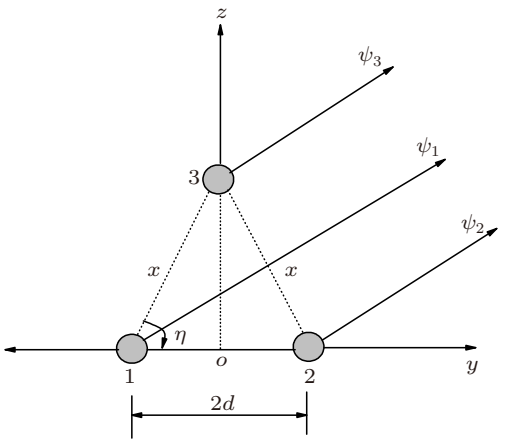


Fig. 1. The schematic diagram of the non-collinear three-center system. The two centers are on y axis separated by inter atomic distance $2d$, while the third center is on z axis separated by distance x from each center, x can be changed with changing angle η. The laser is polarized along the z direction. The screen is placed at very large distance from the system. The distance between 1 and 3 or 2 and 3 can be varied by the equation $x = d\sec\eta$.

Let ψ_1, ψ_2, and ψ_3 be the detached-electron waves from the centers 1, 2, and 3 respectively as shown in Fig. 1. Following the previous approach as in Ref. [16], the resultant outgoing detached-electron wave $\psi_{\rm D}$ can be written as linear combination of individual waves as

$$\psi_{\rm D} = \frac{1}{\sqrt{3}}\sum_1^3 c_i\psi_i, \tag{2}$$

where c_i $(i = 1,2,3)$ are the normalization constants of our system and can be calculated using the molecular orbit theory. In case of homo-nuclear tri-atomic negative ion, $c_i = 1$ for all

$i = 1, 2, 3$. Let (r_i, θ_i, ϕ_i) represent the spherical coordinates of the detached electron relative to each center. Therefore the outgoing detached-electron wave generated from each is given by[7]

$$\psi_i = \frac{4ik^2B}{(k_b^2+k^2)^2}\cos\theta_i \frac{\exp(ikr_i)}{kr_i} \quad \text{for} \quad i = 1,2,3. \tag{3}$$

In Eq. (3) $k = \sqrt{2E}$ and E is the detached-electron energy. By substituting Eq. (3) into Eq. (2), we get the explicit expression for the detached-electron wave function. We can further simplify this expression by applying the large distance approximation that is the distances r_i are much larger than the interatomic distances of our system. Let (r, θ, ϕ) be spherical coordinates of the detached-electron with respect to the origin. Then we approximate the phase terms in Eq. (3) using

$$\begin{aligned} r_1 &\approx r + d\sin\theta\sin\phi, \\ r_2 &\approx r - d\sin\theta\sin\phi, \\ r_3 &\approx r - d\cos\theta\tan\eta. \end{aligned}$$

All other places we can set $r \approx r_1 \approx r_2 \approx r_3$ and $\theta_1 \approx \theta_2 \approx \theta_3 \approx \theta$. With these approximations, the detached-electron wave function ψ_D in Eq. (2) becomes

$$\psi_D = \frac{1}{\sqrt{3}}\frac{4ik^2B}{(k_b^2+k^2)^2}\cos\theta[\exp(-iF) + 2\cos(kd\sin\theta\sin\phi)] \times \frac{\exp(ikr)}{r}. \tag{4}$$

Here we denote $F = kd\cos\theta\tan\eta$, which is a dimensionless quantity, describes the shape of molecule. For $\eta = 0°$, i.e., for collinear shape of the molecule, we get $F = 0$, then the wave function in Eq. (4) reduces exactly to the wave function as given in Ref. [16] (Eq. (3) there in).

In order to find the detached-electron flux in the radial direction for the non-collinear tri-atomic negative ion, the wave function obtained in Eq. (3) is treated quantum mechanically with the following relation

$$J_r(r,\theta,\phi) = j\cdot\hat{r} = \frac{i}{2}(\psi_D\nabla\psi_D^* - \psi_D^*\nabla\psi_D)\cdot\hat{r}. \tag{5}$$

The relation for flux in the radial direction $(J_r(r,\theta,\phi) = J(r,\theta,\phi))$ is given by

$$J(r,\theta,\phi) = J_0(r,\theta)\frac{1}{3}\left[1 + 4\cos(kd\sin\theta\sin\phi)\cos(F) + 4\cos^2(kd\sin\theta\sin\phi)\right]. \tag{6}$$

In Eq. (6),

$$J_0(r,\theta) = k\left(\frac{4ik^2B}{(k_b^2+k^2)^2}\right)^2\frac{\cos^2\theta}{r^2}$$

is the electron flux in the radial direction for H^- as a one center system.

The detached electron flux at any point $\rho = \sqrt{x^2+y^2}$ on the screen placed in the XY-plane at a distance L from the system is

$$J_z(\rho) = J_0(\rho)\frac{1}{3}\left[1 + 4\cos\left(\frac{kdy}{\sqrt{\rho^2+L^2}}\right)\cos\left(\frac{kdL\tan\eta}{\sqrt{\rho^2+L^2}}\right) + 4\cos^2\left(\frac{kdy}{\sqrt{\rho^2+L^2}}\right)\right], \tag{7}$$

with

$$J_0(\rho) = k\left(\frac{4ik^2B}{(k_b^2+k^2)^2}\right)^2\frac{L^3}{(\rho^2+L^2)^{5/2}}.$$

3. Results and discussion

Our result in Eq. (7) can be reduced to collinear form by putting $\eta = 0°$ in it, the second term reduces to $4\cos(kdy/\sqrt{\rho^2+L^2})$. So the formula in Eq. (7) becomes exactly the same as for the collinear tri-atomic negative ion in Ref. [16] (Eq. (9) therein). By using Eq. (7), we can calculate the electron flux distributions of a non-collinear tri-atomic negative ion on a given observation plane for different energies $E = 30$, 150, 300 eV as shown in the figures (Figs. 2–4). For fixed energy $E = 30$ eV with different geometrical shapes of the molecule for $\eta = 60°$, 45°, 30°, 0°, we plot the result in Fig. 2. From Fig. 2, we find that the electron flux distribution oscillates with different energies. By increasing the energy, the central point also increases for different geometrical shapes. Where η-value defines the shape of the given molecular system. From the figures we can see that this η-value has a great influence on the electron flux distribution. Figures 2(d), 3(d), and 4(d) show the case when $\eta = 0°$. Under this condition, the molecule looks like a collinear molecule.[16] These oscillations in the electron flux are because of the interference of three trajectories each coming from each center of the non-collinear molecule. As the detached electron energy increases, the electron flux is shown to exhibit the damping oscillation as is clearly seen in the Fig. 4. The clear picture of interference of tri-atomic anion for fixed energy $E = 30$ eV with different geometrical shapes of the molecule for $\eta = 60°$, 45°, 30°, 0° is shown in three dimensions in Fig. 5.

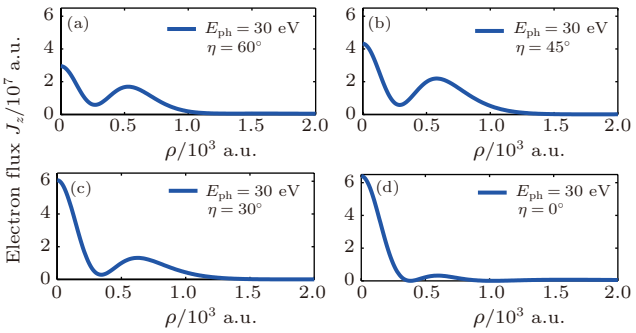


Fig. 2. (color online) The detached electron flux given as in Eq. (6), is plotted versus ρ. The energy of $E_{ph} = 30$ eV, $L = 1000$ a.u., $d = 4$ a.u. but the geometry of the non-collinear molecule is different. At $\eta = 0°$, we obtained the same result as in Ref. [16].

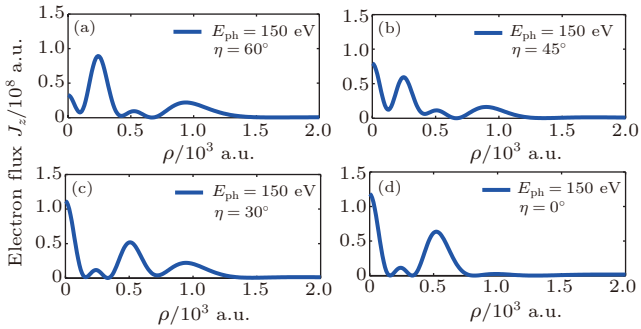


Fig. 3. (color online) The detached-electron flux with energy $E_{ph} = 150$ eV, $L = 1000$ a.u., $d = 4$ a.u. for different η-values.

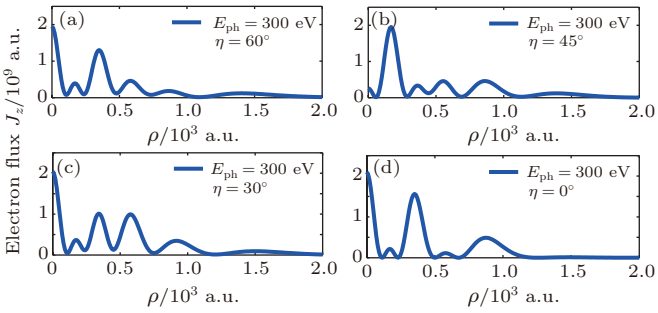


Fig. 4. (color online) The detached-electron flux with energy $E_{ph} = 300$ eV, $L = 1000$ a.u., $d = 4$ a.u. for different η-values.

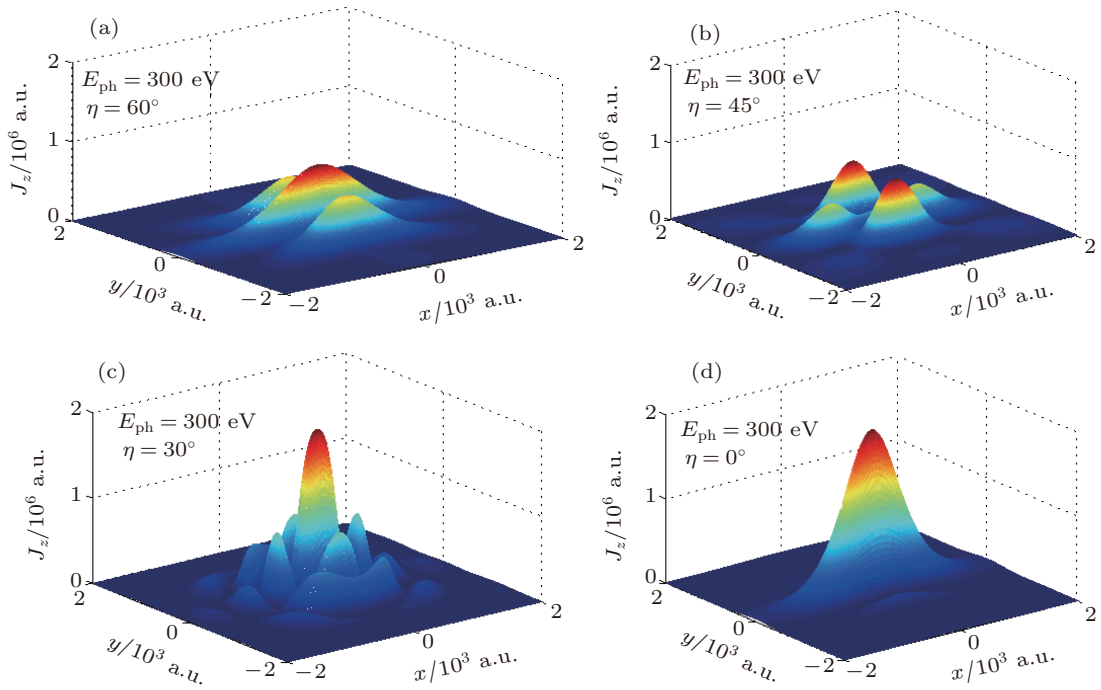


Fig. 5. (color online) The detached electron flux on the screen in three dimensions for the non-collinear tri-atomic negative ions. In the sketch, we set $E_{ph} = 30$ eV, $d = 4$ a.u., $L = 1000$ a.u. While the values of the angle η are 60°, 45°, 30°, and 0°. The peaks due to interference of detached electron for the three center non-collinear system are clearly visible.

4. Conclusion

In conclusion, we have studied the photo-detachment of non-collinear tri-atomic homo-nuclear anion using a three-center model, in particular the influence of geometrical shape on the electron flux distribution was discussed. We have derived a general relation for the detached-electron flux distribution for z-polarized laser light. The results show that the geometrical shape has an important effect on the electron flux distributions. The interference in the electron flux is because of three different trajectories originating from each atom of the system. When they reach a sensitive detector with different phases, the interference phenomena occur. Our study suggests that we can estimate the shape of the molecule by analyzing the photo-detachment spectra. The oscillations in the present non-collinear three-center system can be explained using closed orbit theory as interference between detached electron waves produced from one center and the sources at the other centers. We hope that this model may benefit the understanding of the non-collinear tri-atomic molecule in photo-detachment microscopy experiments.

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