Oscillations in the total photodetachment cross sections of a linear triatomic negative ion

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The total photodetachment cross section of a linear triatomic negative ion using plane polarized laser light parallel to the axis of the molecular ion is derived. The cross section shows strong oscillations in contrast to the recently reported case, where the laser polarization was perpendicular to the axis of the triatomic molecular ion (Appl. Phys. Lett. 94, 041125, 2009). Closed orbit theory is used to explain these oscillations and the results are compared with a diatomic molecule.

I. INTRODUCTION

Interest in the photodetachment process and photodetachment spectroscopy has grown in the recent years [1–12]. The photodetachment cross section for hydrogen negative ion has been investigated experimentally [13, 14] and theoretically [15–23]. The most intrusting aspect of the H− cross section in the presence of a static electric field of a few hundred kV/cm above threshold, it shows oscillations while smooth in the absence of the field. Afaq and Du extended one-center model and developed two-center model for the photodetachment of electron from a diatomic molecule [2, 4, 5]. They gave the idea of special detached-electron orbit starting from one atom and ending at the other atom in a diatomic negative molecule. This special detached-electron orbit is responsible for the oscillations in the total photodetachment cross sections and is the updated version of the closed orbit theory [19, 24, 25].

Recently Afaq et al [1] showed a smooth behavior of the total photodetachment cross section of a linear triatomic negative ion placed perpendicular to the direction of polarization of the laser beam. The total cross section approaches one-center and two-center models for a highly energetic
laser beam while it approaches to one-center model for a low energy laser. In the present article we have investigated the photodetachment cross section for a linear triatomic negative ion with the laser beam polarized parallel to the axis of the triatomic ion. A procedure similar to the one reported by Afaq et al. is used in the present calculation.

Three centers of the linear triatomic negative ion is supposed as a coherent source of electrons similar to H\(^-\) and the detached-electron wave function is obtained as a superposition of the three coherent waves originating from each center of the system. The detached-electron flux, due to the superposition of these three waves, on a screen is calculated. For the total photodetachment cross section, of a linear triatomic negative ion, an analytical formula is derived by the integration of the electron flux in all directions. Our present article will be helpful in the understanding of the structural information of a linear triatomic negative ions like BeCl\(_2\), HCN\(^-\), CS\(_2\), CO\(_2\) etc. Atomic units are used throughout the article or mentioned otherwise.

II. DETACHED ELECTRON WAVE FUNCTION

The schematic diagram of the linear triatomic negative ion as a three-center model is shown in Fig.1. Numbers 1, 2 and 3 on the z axis represent the three centers of the system for the negative molecular ion, such that number 3 is set as the origin of the coordinates system. The ± symbols indicate the sign of the two lobes of the p-orbital wave function. The screen is placed at a distance \(L\) from the three-center system and perpendicular to the z axis. Assume \(d\) is the distance between the two adjacent centers and is of the order of a few atomic units, while \(L\) is much larger than \(d\) and is equal to several thousands atomic units. These two variables can be changed in our model.

Let there is only one active electron like the H\(^-\) model. The normalized wave function for the active electron in the three-center system is the linear superposition of the H\(^-\) like bound state at the three centers, \(\phi_T = (\phi_1 + \phi_2 + \phi_3)/\sqrt{3}\). Where \(\phi_1\), \(\phi_2\) and \(\phi_3\) are the normalized wave functions for H\(^-\) but centered at 1, 2 and 3 in Fig. 1. Photodetachment process is a two step process. In the first step, a negative ion absorbs one photon energy \(E_{ph}\) and generates an outgoing electron wave, while in the second step, this outgoing wave propagates to a large distance.

A z-polarized laser light similar to Ref. is used. The detached-electron wave function can be obtained by the linear superposition of the three coherent waves generated from the each center. These coherent waves can be achieved from the results of the H\(^-\) photodetachment in the absence of an electric field. Assume \(\Psi_1^+, \Psi_2^+\), and \(\Psi_3^+\) are the waves emitted from centers 1, 2 and 3.
The detached electron wave function $\Psi^+_M$ is given by:

$$\Psi^+_M = \frac{1}{\sqrt{3}}(\Psi^+_1 + \Psi^+_2 + \Psi^+_3) \quad (1)$$

Spherical polar coordinates $(r_1, \theta_1, \phi_1)$, $(r_2, \theta_2, \phi_2)$ and $(r_3, \theta_3, \phi_3)$ for the detached-electron relative to each center are used. The emitted waves, $\Psi^+_1$, $\Psi^+_2$ and $\Psi^+_3$, can be expressed using Ref. [20] by:

$$\Psi^+_1(r_1, \theta_1, \phi_1) = C \cos \theta_1 \frac{\exp(ikr_1)}{kr_1}$$

$$\Psi^+_2(r_2, \theta_2, \phi_2) = C \cos \theta_2 \frac{\exp(ikr_2)}{kr_2}$$

$$\Psi^+_3(r_3, \theta_3, \phi_3) = C \cos \theta_3 \frac{\exp(ikr_3)}{kr_3},$$

where $C = \frac{4kB_i}{(k^2 + k^2)b^2}$, $k_b$ is related to the binding energy $E_b$ of H$^-$ by $E_b = \frac{k^2}{2}$, and B is a normalization constant, having value of 0.31552.

The detached-electron wave function $\Psi^+_M(r, \theta, \phi)$ from the three-center system can be obtained after the substitution of each center wave function in Eq. (1). The problem can be further simplified by using the idea that $L$ is much larger than $d$. Let $(r, \theta, \phi)$ be the spherical coordinates of the detached electron. For the phase terms, $r_1 \approx r - d \cos \theta$, $r_2 \approx r + d \cos \theta$ and $r_3 \approx r$ are used, while in all other places $r_1 \approx r_2 \approx r_3 \approx r$ and $\theta_1 \approx \theta_2 \approx \theta_3 \approx \theta$. With these approximations $\Psi^+_M(r, \theta, \phi)$ reduces to:

$$\Psi^+_M(r, \theta, \phi) = \frac{C \cos \theta}{\sqrt{3}} \left[ 1 + 2 \cos(kd \cos \theta) \right] \frac{\exp(ikr)}{r} \quad (2)$$

### III. DERIVATION OF THE TOTAL CROSS SECTION FOR THE TRIATOMIC NEGATIVE ION

The detached-electron flux in the radial direction for the triatomic negative ion is calculated as in [1]:

$$j_r(r, \theta, \phi) = \frac{kC^2 \cos^2 \theta}{3r^2} \left[ 1 + 4 \cos(kd \cos \theta) + 4 \cos^2(kd \cos \theta) \right] \quad (3)$$

A large imaginary spherical surface $\Gamma$ enclosing the triatomic negative ion is used to investigate the behavior of the total cross section. From the electron flux crossing the surface a generalized differential cross section $\frac{d\sigma(q)}{ds}$ can be obtained [20]:

$$\frac{d\sigma(q)}{ds}$$
\[
\frac{d\sigma(q)}{ds} = \frac{2\pi E_{ph}}{c} \vec{j_r} \cdot \hat{n}
\]  

(4)

where \(q\) is the coordinate on the surface \(\Gamma\), \(\hat{n}\) is the exterior norm vector at \(q\), \(c\) is the speed of light in a.u. and \(ds = r^2 \sin \theta d\theta d\phi\) is the differential area on the spherical surface. The total cross section of the triatomic negative ion can be derived by the integration of the differential cross section over the entire surface, \(\sigma(q) = \int_{\Gamma} \frac{d\sigma(q)}{ds} ds\). Substituting the value of \(\vec{j_r}\) from Eq. (3) in Eq. (4) and integrating:

\[
\sigma(E) = \frac{2\pi k C^2 E_{ph}}{3c} \int_0^\pi \int_0^{2\pi} \cos^2 \theta \sin \theta [1 + 4 \cos(kd \cos \theta) + 4 \cos^2(kd \cos \theta)] d\theta d\phi
\]

After solving the integration, the following simple result is obtained:

\[
\sigma(E) = \sigma_0(E) A_{||}(kd)
\]

(5)

where

\[
\sigma_0(E) = \frac{8\pi^2 k |C|^2 E_{ph}}{3c} \quad \text{with} \quad C = \frac{4kBi}{(k^2 + k^2)^2}
\]

(6)

\[
A_{||}(kd) = 1 + 4 \left[ \frac{\sin(kd)}{(kd)} + 2 \frac{\cos(kd)}{(kd)^2} - 2 \frac{\sin(kd)}{(kd)^3} \right] + 2 \left[ \frac{\sin(2kd)}{(2kd)} + \frac{\cos(2kd)}{(2kd)^2} - \frac{\sin(2kd)}{(2kd)^3} \right]
\]

(7)

\(\sigma_0(E)\) is the smooth total photodetachment cross section of \(H^-\) and \(A_{||}(kd)\) is the modulation function due to the triatomic negative ion. The total photodetachment cross-section expressed in Eq. (5) is plotted in Fig. 2 as a function of the incident laser photon energy (eV) for several \(d\) values. The figure shows strong oscillation for the laser polarization parallel to the axis of the linear triatomic molecule, in contrast to the recently reported \[1\] laser polarization perpendicular to the triatomic molecule. Closed orbit theory \[19, 24, 25\] is used to explain the origin of these oscillations. The detached-electron wave produced at center 1 while propagates out of the source reaches to center 3 and center 2 on its way out. The overlap of the detached-electron wave from center 1 with the source at center 3 and the source at the center 2 produces oscillations in the total cross section of the triatomic negative ion. As a result we encounter two special orbits of the detached-electron; one is generated from center 1 to the nearby center 3 and the other one is from center 1 to center 2. These two special orbits of the detached-electron are responsible for the oscillations. Similar oscillations are also reported in the diatomic negative ion (two-center system) \[2\]. A comparison of our results with the two-center model is shown in Fig. 3. Fourier transformation in Fig. 3 is used to investigate special orbits of the detached-electron.
IV. CONCLUSION

A plane polarized laser, polarized parallel to the axis of the linear tri-atomic negative ion, is used to investigate the behavior of the detached electron from a triatomic negative ion. A simple analytical formula for the total cross section is obtained which shows strong oscillations in contrast to the case reported in [1]. Closed orbit theory is used to explain these oscillations as the interference between detached-electron waves produced from one center and sources at the other two centers. Fourier transformation in Fig. 3 shows that these two types of orbits are responsible for the oscillations in the total photodetachment cross section and hence there are two oscillation frequencies in the three-center system in contrast to only one oscillation frequency in the two-center system. Our theoretical study suggests that the photodetachment of negative ion as a three-center model is interesting, and the interference can be used to investigate structural information of linear triatomic negative ions. However this model can optimally be modified for non-linear triatomic ions.

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Figure captions

**Fig.1**  Schematic diagram of the three-center model as a triatomic negative ion

**Fig.2**  The total photodetachment cross section for different values of $d$ using Eqs. (5)-(7). From (a) to (d) show that with the increase in $d$ the oscillation amplitude decreases and oscillation frequency increases

**Fig.3**  Fourier transformation of the total photodetachment cross section (a) calculation, (b) comparison: the dashed line is the result with two-center system and solid line is the result with three-center system. The calculations are carried out using $d = 250$ a.u.
(a) $d = 250 \, a_0$

(b) $|F(D)| \times 10^{-3} \, \text{a.u.}$

- Three-center Model
- Two-center Model