Oscillations in the total photodetachment cross sections of a triatomic anion

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(Dated: January 6, 2010)

Abstract

The total photodetachment cross section of a linear triatomic anion is derived for arbitrary laser polarization direction. The cross section is shown to be strongly oscillatory when the laser polarization direction is parallel to the axis of the system; the oscillation amplitude decreases and vanishes as the angle between the laser polarization and the anion axis increases and becomes perpendicular to the axis. The average cross section over the orientations of the triatomic system is also obtained. The cross section of the triatomic anion is compared with the cross section of a two-center system. We find there are two oscillation frequencies in the triatomic anion in contrast to only one oscillation frequency in the two-center case. Closed-orbit theory is used to explain the oscillations.

PACS numbers: 32.80.Fb,32.80.Qk,33.70.Ca
I. INTRODUCTION

Photodetachment of negative ions in the presence of a static electric field has been an active research area in the last decades [1–25]. The most interesting feature in the cross section is the induced oscillations above photodetachment threshold by the static electric field. The oscillations in the total cross sections can be understood using closed-orbit theory [26–28]. In an effort to understand the oscillations in various processes involving two-center system such as the photoionization cross section of diatomic molecule [29], the scattering of $D_2$ molecule by fast electron [30], a molecule in a strong laser field [31], above-threshold ionization [32] and harmonic generation [33], Afaq and Du extended the one-center $H^-$ model for photodetachment and developed a two-center model for photodetachment [34–36]. They demonstrated the cross sections in the two-center system show strong oscillation which can be explained using closed-orbit theory. In particular, a detached electron orbit connecting the two centers is identified to be responsible for the oscillation in the total photodetachment cross sections of the two-center system.

To understand the structural information on linear triatomic negative anions such as $\text{BeCl}_2^-$, $\text{HCN}^-$, $\text{CS}_2^-$ and $\text{CO}_2^-$ [37], Afaq et al. [38] recently studied the photodetachment of a triatomic anion with three centers when the axis of the triatomic ion is perpendicular to the laser polarization direction. Interference patterns for detached-electron on a screen placed at a large distance from the system were demonstrated, but the total cross section was found to be smooth and no oscillation was observed for this configuration.

Here we extend the study of the total photodetachment cross section of the above triatomic anion system to the general case with an arbitrary laser polarization direction. We will derive analytic formulas for the total cross section which depends on photon energy, laser polarization direction and other parameters characterizing the triatomic anion. It will be shown that the cross section shows strong oscillations when the laser polarization is parallel to the system axis and the oscillation amplitudes gradually decrease to zero as the laser polarization is changed to be perpendicular to the axis. We also obtained the cross section averaged over the orientations of the system. We compare the cross section of the triatomic anion with that of the two-center system. The oscillations in photodetachment cross sections for the triatomic anion appear much enhanced compared to the two-center case. We find there are two oscillation frequencies in the triatomic anion. The two oscillations are
explained using closed-orbit theory. Atomic units will be used unless specially noted.

II. FORMULAS FOR TOTAL PHOTODETACHMENT CROSS SECTION

The linear triatomic anion interacting with a laser is shown schematically in Fig.1. Symbols 1,0 and 2 represent the three atomic centers in the system. It is convenient to choose the z-axis in the direction of the three-center axis and the middle center denoted by 0 as the origin of coordinates. Let $d$ be the distance between two adjacent centers. The laser polarization direction is denoted as $(\theta_L, \phi_L)$ with respect to the z axis.

In the triatomic anion, one active electron is assumed. This is an extension of the two-center model for photodetachment\cite{34–36} to a three-center model by Afaq et al.\cite{38}. In the photodetachment process, there are two steps: in the first step, the active electron absorbs one photon energy $E_{ph}$ and escapes from the negative anion as an electron wave from each center; in the second step, the outgoing waves from each center propagate out to large distances. The interference of the outgoing waves from each center produces oscillatory cross section.

For the general case, as illustrated in Fig. 1, Let $\Psi_1^+, \Psi_0^+$ and $\Psi_2^+$ be the detached-waves from center 1, 0 and 2 respectively. Following the previous approach in the two-center case\cite{36}, the outgoing detached-electron wave $\Psi_M^+$ from the triatomic anion can be written as a linear combination given by\cite{36}

$$\Psi_M^+ = \frac{1}{\sqrt{3}}(\Psi_1^+ + \Psi_0^+ + \Psi_2^+).$$

(1)

Let $(r_1, \theta_1, \phi_1)$, $(r_0, \theta_0, \phi_0)$ and $(r_2, \theta_2, \phi_2)$ represent the spherical coordinates of the detached-electron relative to the three centers respectively. The detached-electron wave generated from each center has been worked previously\cite{25}. They can be written as

$$\Psi_1^+ = \frac{4Bk^2i}{(k_b^2 + k^2)^2}f(\theta_1, \phi_1; \theta_L, \phi_L)\frac{\exp(ikr_1)}{kr_1},$$

$$\Psi_0^+ = \frac{4Bk^2i}{(k_b^2 + k^2)^2}f(\theta_0, \phi_0; \theta_L, \phi_L)\frac{\exp(ikr_0)}{kr_0},$$

$$\Psi_2^+ = \frac{4Bk^2i}{(k_b^2 + k^2)^2}f(\theta_2, \phi_2; \theta_L, \phi_L)\frac{\exp(ikr_2)}{kr_2},$$

(2)

where $k = \sqrt{2E}$ and $E$ is the detached-electron energy; $k_b$ is related to the binding energy $E_b$ of H$^-$ by $E_b = \frac{k_b^2}{2}$. The photon energy is given by $E_{ph} = E + E_b$. $B$ is a normalization
constant $c$, $c$ is the speed of light and is approximately equal to 137 a.u.. The angular factor such as $f(\theta_0, \phi_0; \theta_L, \phi_L)$ represents the dependence of the detached-electron wave function on the outgoing direction $(\theta_0, \phi_0)$ for center 0, and it can be written as

$$f(\theta_0, \phi_0; \theta_L, \phi_L) = \cos \theta_0 \cos(\theta_L) + \sin \theta_0 \sin(\theta_L) \cos(\phi_0 - \phi_L).$$

(3)

The angular factors $f(\theta_1, \phi_1; \theta_L, \phi_L)$ and $f(\theta_2, \phi_2; \theta_L, \phi_L)$ can be written out in a similar way.

After substituting Eqs.(2) and Eqs.(3) in Eq.(1), we can get the explicit expression for the detached-electron wave function $\Psi_M^+$. The resulting formula for $\Psi_M^+$ can be simplified because the calculation of the photodetachment cross section requires the knowledge of $\Psi_M^+$ at large distances where $r_1$, $r_0$ and $r_2$ are much greater than the distance $d$ between two neighboring centers. Let $(r, \theta, \phi)$ be the spherical coordinates of the detached-electron relative to the origin. Then we approximate the phase terms using $r_1 \approx r - d \cos \theta$, $r_0 = r$, $r_2 \approx r + d \cos \theta$; in other places we can set $r_1 \approx r_2 \approx r_0 = r$, $\theta_1 \approx \theta_2 \approx \theta_0 = \theta$. With these approximations, $\Psi_M^+$ becomes

$$\Psi_M^+(r, \theta, \phi) = \frac{4Bk^2i}{(k_0^2 + k^2)^2} \frac{1}{\sqrt{3}} f(\theta, \phi; \theta_L, \phi_L)[1 + 2 \cos(kd \cos \theta)] \exp(ikr) \frac{kr}{kr}, r \rightarrow \infty.$$  

(4)

Eq.(4) describes the detached-electron wave of the linear triatomic anion when the detached-electron is far away from the anion.

The differential cross section is

$$\frac{d\sigma(q)}{ds} = \frac{2\pi E_{ph}}{c} j \cdot n, \quad (5)$$

$$j = \frac{i}{2}(\Psi_M^+ \nabla \Psi_M^+ - \Psi_M^{++} \nabla \Psi_M^{++}),$$

where $\sigma$ is the photodetachment cross section, $ds$ is the differential area on a surface $\Gamma$ such as the surface of a large sphere enclosing the anion, $q$ is the coordinate on the surface $\Gamma$, $n$ is the exterior norm vector at $q$ and $j$ is the detached-electron flux.

The detached-electron flux in the radial direction can be evaluated as

$$j_r(r, \theta, \phi) = j \cdot \hat{r} = \frac{i}{2}(\Psi_M^+ \partial \Psi_M^+ - \Psi_M^{++} \partial \Psi_M^{++}).$$

(6)

When Eq.(4) is substituted in Eq.(6), we get

$$j \cdot \hat{r} = \frac{16B^2k^4}{3k(k_0^2 + k^2)^2} \frac{f^2(\theta, \phi; \theta_L, \phi_L)}{r^2} [1 + 4 \cos(kd \cos \theta) + 4 \cos^2(kd \cos \theta)].$$

(7)
To get the total photodetachment cross section, we integrate the differential photodetachment cross section using Eq.(5) and Eq.(7) over $\theta$ and $\phi$,

$$\sigma(E, d, \theta_L) = \frac{2\pi E_{ph} c}{r^2} \int (\mathbf{j} \cdot \hat{r}) r^2 \sin \theta d\theta d\phi. \quad (8)$$

After a straightforward integration, we find the total photodetachment cross section can be written as a product form

$$\sigma(E, d, \theta_L) = \sigma_0(E) A_3(kd, \theta_L), \quad (9)$$

where

$$\sigma_0(E) = \frac{16\sqrt{2} B^2 \pi^2 E^4}{3c(E_b + E)^3}, \quad (10)$$

$$A_3(kd, \theta_L) = 1 + \frac{4}{3} I(kd) + \frac{2}{3} I(2kd). \quad (11)$$

In the above equations, $\sigma_0(E)$ is the smooth total photodetachment cross section of $\text{H}^-[3]$ and $A_3(kd, \theta_L)$ is a modulation function for the triatomic anion. The function $I(S)$ appearing in Eq.(11) is given by

$$I(S) = 3 \cos^2 \theta_L \left[ \frac{\sin S}{S} + 3 \frac{\cos S}{S^2} - 3 \frac{\sin S}{S^3} \right] + 3 \left[ \frac{\sin S}{S^3} - \frac{\cos S}{S^2} \right]. \quad (12)$$

The function $I(S)$ also appears in the modulation function for the two-center problem studied previously [36].

III. OSCILLATIONS AND LIMITS OF CROSS SECTIONS

First we show the cross section obtained by Afaq et al. [38] for the perpendicular configuration can be obtained from the general formulas. When the laser polarization direction is perpendicular to the direction of the axis, $\theta_L = \frac{\pi}{2}$. From Eqs.(9)-(12) we immediately have

$$\sigma(E, d, \frac{\pi}{2}) = \sigma_0(E)[1 + 4\left( \frac{\sin(kd)}{(kd)^3} - \frac{\cos(kd)}{(kd)^2} \right) + 4\left( \frac{\sin(2kd)}{(2kd)^3} - \frac{\cos(2kd)}{(2kd)^2} \right)]. \quad (13)$$

The result in Eq.(13) is identical to that given by Afaq et al. [38]. In Fig.2 we compare the photodetachment cross section in Eq.(13) with the cases $\theta_L = 0$ and $\theta_L = \frac{\pi}{4}$ in Eqs.(9)-(12) when $d = 4a_0$. Indeed we do not find any oscillation in the the cross section for $\theta_L = \frac{\pi}{2}$ in agreement with Afaq et al. [38]. But when the direction of laser polarization is
not perpendicular to the axis, we observe a hint of oscillation as illustrated in the insert. In fact, the oscillations will become strong and obvious as the parameter $S = kd$ is increased to be comparable or greater than $\pi$. In Fig.3 we show the photodetachment cross sections in Eqs.(9)-(12) for several $d$ and $\theta_L$ values. The following points can be derived directly from Fig.3. First, we do not observe obvious oscillation in the photodetachment cross sections for the perpendicular laser polarization ($\theta_L = \frac{\pi}{2}$) even when $d$ is increased. But when the laser polarization direction is not perpendicular to the anion axis, there are strong oscillations. Second, the oscillation amplitude depends on the laser polarization direction. In fact, the oscillation amplitude increases and reaches maximum as the laser polarization direction is turned from perpendicular to parallel direction with respect to the anion axis ($\theta_L = 0$). However, the oscillation frequency is not sensitive to the laser polarization direction. The modulation function $A_3$ suggests the oscillation frequency increases but the oscillation amplitude decreases as the parameter $d$ is increased. The oscillations are easily observed when $d$ is approximately around $30 - 100a_0$ in our system.

The limits of the photodetachment cross sections of the anion can be obtained from the limits of $I(S)$. Using Taylor series expansion, one can show

\[
\lim_{S \rightarrow 0} I(S, \theta_L) = 1, \\
\lim_{S \rightarrow \infty} I(S, \theta_L) = 0.
\]

The above results are independent of the value of $\theta_L$. Therefore we conclude from Eqs.(9)-(12) that in the low energy limit the total photodetachment cross section of the three-center system is three times of the photodetachment cross section of a single-center system and in the large photon energy limit the photodetachment cross section of the three-center system approaches the photodetachment cross section of a single-center system.

It is also straightforward to get the photodetachment cross section averaged over the orientations of the anion. Let us assume the direction of the anion is random with respect to the laser polarization direction. Defining the averaging by

\[
\bar{\sigma}(E, d) = \frac{1}{\int_0^\pi \sin \theta_L d\theta_L \int_0^{2\pi} \sigma(E, d, \theta_L) d\phi_L / \int_0^\pi \sin \theta_L d\theta_L \int_0^{2\pi} d\phi_L}.
\]

The integrals can be evaluated to give the following results,

\[
\bar{\sigma}(E, d) = \sigma_0(E) \bar{A}_3(kd), \\
\bar{A}_3(kd) = 1 + \frac{4 \sin(kd)}{3kd} + \frac{2 \sin(2kd)}{3 \cdot 2kd}.
\]
It is interesting that the average cross section is equal to the laser polarization dependent cross section in Eqs.(9)-(12) evaluated at a special angle satisfying $\theta_L = \arccos\left(\frac{1}{\sqrt{3}}\right)$ (approximately 54.74°). The same angle was noticed for the two-center system earlier.

**IV. FOURIER ANALYSIS**

The averaging process does not change the basic oscillatory structure of the cross section. However, the oscillation amplitudes in the average cross section are reduced to one third of the values obtained for the laser polarization dependent cross section at $\theta_L = 0$. In Fig.4(a) we compare the average cross section and the laser polarization dependent cross section at $\theta_L = 0$.

The oscillations are better analyzed using a transformation. For the cross section $\sigma(E)$ we define $F(x)$ using the following integral

$$F(x) = \int_{E_1}^{E_2} [\sigma(E) - \sigma_0(E)] \sin\left[\pi \left(\frac{E - E_1}{E_2 - E_1}\right)\right] k \exp(-ikx) dE. \quad (18)$$

In Fig.4(b) we present the corresponding transformations of the two cross sections in Fig.4(a). Several points can be derived from Fig.4 regarding the oscillations. First, there are two peaks in each transformation. The peaks correspond to two oscillations in each cross section. Second, the oscillation frequencies are not changed by the averaging procedure. Third, the oscillation amplitudes in the average cross section are considerably reduced.

We now compare the average photodetachment cross section of the triatomic anion with that of the two-center negative ion studied earlier. In Fig.5(a) we show the two cross sections. One can see that the oscillation in the photodetachment cross section for the triatomic anion is enhanced compared to that for the two-center system. In Fig.5(b) we show the corresponding transformations of the two cross sections in Fig.5(a). It is clear from Fig.5(b) that there are two oscillations for the triatomic anion but only one oscillation for the two-center system.

Closed-orbit theory was extended previously to explain the oscillation in the cross section of the two-center system. The oscillation was identified as an interference between the detached-electron wave emitted at one center and the source of the wave at another center. A similar explanation can be made here. For example, the detached-electron wave produced at center 1 will reach center 0 and 2 as it propagates out. The overlap of this
detached-electron wave from center 1 with the source at center 0 and the source at 2 produce the \( \sin(kd) \) and \( \sin(2kd) \) oscillations in the total cross section. Detached-electron wave from center 2 also have similar interference terms. The final oscillation amplitudes include all the interference terms. We will not repeat the detail of such derivation which is quite similar to the two-center case\[36\]. We emphasize that such a derivation based on closed-orbit theory establishes that \( kd \) is the action \( \int \mathbf{p} \cdot d\mathbf{q} \) of the detached-electron propagating from one center to a neighboring center and \( 2kd \) is the action of the detached-electron propagating from center 1 to center 2 or from center 2 to center 1. The two oscillations are directly associated with the detached-electron orbits from one center to a neighboring center having action \( kd \) and the detached-electron orbits connecting center 1 and 2 having action \( 2kd \).

V. CONCLUSIONS

We have studied the photodetachment of the recent triatomic anion model\[38\] in the general case. We have derived the total photodetachment cross section for arbitrary laser polarization direction. It is demonstrated there are two oscillation frequencies in the cross sections. The amplitudes of the oscillations can be varied by changing the laser polarization direction. The amplitudes are largest when the laser polarization is parallel to the anion axis. As the laser polarization direction is turned to be perpendicular to the axis, the oscillation amplitudes decrease and vanish. We also obtained the average cross section for random orientations of anion. The averaging procedure modifies the oscillation amplitudes but it does not change the oscillation frequencies. The two oscillations in the present three-center system can be explained using closed-orbit theory as interference between detached-electron waves produced from one center and the sources at other centers. Two types of detached-electron orbits are responsible for the two oscillations.

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FIG. 1: The schematic representation for the photodetachment of the linear triatomic anion. The dotted arrows point to the laser polarization direction. The plus and minus lobes represent the angular amplitude of the detached-electron from each center.
FIG. 2: The photodetachment cross section of the triatomic anion at $d = 4a_0$ and the laser polarization direction $\theta_L$ are respectively equal to 0, $\pi/4$, $\pi/2$. The inset provides a hint of oscillation for $\theta_L = 0$ and $\theta_L = \pi/4$. 
FIG. 3: The photodetachment cross sections of the triatomic anion with different values of $d$ and $\theta_L$. 
FIG. 4: (a) The total photodetachment cross section for $d = 100a_0$ and $\theta_L = 0$ (solid line) and the average cross section (dotted line). (b) The transformations defined in Eq.(18) for the above two cross sections.
FIG. 5: (a) The orientation average photodetachment cross sections of the two-center system and the orientation average triatomic anion system. The parameter $d = 100 a_0$. (b) The transformations defined in Eq.(18) for the above two cross sections.