Asymptotic form of the high energy photoionization cross section of fullerenes

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ABSTRACT

We show that the theoretical predictions on high energy behavior of the photoionization cross section of fullerenes depend crucially on the form of the function $V(r)$ which approximates the fullerene field. The shape of the high energy cross section is obtained without solving the wave equation. The cross section energy dependence is determined by the analytical properties of the function $V(r)$.

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1. Introduction

In this paper we calculate the high energy nonrelativistic asymptotics for the photoionization cross section of the valence electrons of fullerenes C_N. We consider the fullerenes which can be treated approximately as having the spherical shape. The photon carries the energy ω which is much larger than the ionization potential I. We find the leading term of the cross section expansion in terms of $1/\omega$. We keep the photon energy to be much smaller than the electron rest energy mc^2. Here we consider only the ionization of s states. We employ the relativistic system of units in which $\hbar = 1$; $c = 1$ and the squared electron charge $e^2 = \alpha = 1/137$.

The actual potential experienced by the fullerene valence electrons is a multicentered screened Coulomb potential produced by the C^{+4} carbon ions of the fullerene. The ionized electron approaches one of these centers transferring large momentum. The asymptotics of the photoionization cross section $\sigma(\omega)$ in the screened Coulomb field is the same as in the unscreened one [1], i.e. $\sigma \sim \omega^{-7/2}$. Thus we expect the observed asymptotic also to be $\sigma \sim \omega^{-7/2}$.

However one usually uses a model central potential $V(r)$ for description of the field created by the fullerene. For a model poten-

tial the asymptotics may be a different one. We consider spherical fullerene with the radius R and the width of the layer $\Delta \ll R$. The general properties of the potential $V(r)$ are well known–see, e.g. [2]. It is located mostly inside the fullerene layer $R - \Delta/2 \leq r \leq R + \Delta/2$ being negligibly small outside.

In the simplest (or even the oversimplified [3]) version it is just the well potential which is constant inside the layer and vanishes outside. Introducing $R_{2,1} = R \pm \Delta/2$ we can present the potential as

$$V(r) = -V_0\theta(r - R_1)\Big(1 - \theta(r - R_2)\Big); \quad V_0 > 0. \tag{1}$$

Recall that $\theta(x) = 1$ for $x \geq 0$ while $\theta(x) = 0$ for $x < 0$. One often uses the Dirac bubble potential

$$V(r) = -U_0\delta(r - R), \tag{2}$$

in the fullerene studies [4]. Here, as well as in Eqs. (3) and (4) $U_0 > 0$ are the dimensionless constants.

The nowadays calculations are often based on the jellium model [5] (see, e.g. [6,7]). In this approach the charge of the positive core consisting of nuclei and the internal electrons is assumed to be distributed uniformly in the fullerene layer. The field of the positive core is $V(r) = V_1(r)$ at $0 \leq r < R_1$, $V(r) = V_2(r)$ at $R_1 \leq r \leq R_2$, and $V(r) = V_3(r)$ at $r > R_2$ with

$$V_1(r) = const = -U_0\frac{3}{2}\frac{R_2^2 - R_1^2}{R_2^3 - R_1^3};$$

$$V_2(r) = -\frac{U_0}{2(R_2^3 - R_1^3)}\left(3R_2^2 - r^2(1 + \frac{2R_1^3}{r^3})\right);$$

$$V_3(r) = -\frac{U_0}{r}. \tag{3}$$

Sometimes model potentials are determined by analytical functions of r with a sharp peak at $r = R$. The Lorentz bubble potential is

$$V(r) = -\frac{U_0}{\pi}\frac{a}{(r-R)^2 + a^2}. \tag{4}$$

It describes the Dirac bubble potential at $a \to 0$. The Gaussian-type potential

$$V(r) = -\frac{V_0}{\pi}\exp\frac{-(r-R)^2}{a^2}, \tag{5}$$

with $a \approx \Delta \ll R$ was employed in [8].

Strictly speaking our analysis is true for the negative ion C_N^-. However, since there are many valence electrons in the fullerene shell, we expect it to be true for photoionization of the neutral fullerene C_N as well.

As it stands now, the asymptotics for the photoionization cross section is known only for the Dirac bubble potential [10]. Here we demonstrate that the energy behavior of the asymptotic cross section is strongly model dependent. It is determined by the analytical properties of the potential $V(r)$. In Sec. 2 we obtain the general equation for the asymptotics of the photoionization cross section. In Sec. 3 we calculate the asymptotics for the potentials mentioned in Introduction. We analyze the results in Sec. 4.

2. Asymptotics of the cross section

The photoionization cross section can be presented as (see Eq. (56.3) of [9] or Eq. (5.76) of [4])

$$d\sigma = n_e\frac{mp}{(2\pi)^2}|F|^2 d\Omega. \tag{6}$$

Here m is the electron mass, $p = |\mathbf{p}|$, while $\mathbf{p}$ is the photoelectron momentum, Ω is the solid angle of the photoelectron, and n_e is the number of electrons in the ionized state. The normalization factor of the photon wave function $n(\omega) = \sqrt{4\pi}/\sqrt{2\omega}$ is included in the photoionization amplitude F. Averaging over polarizations of the incoming photon is assumed to be carried out.

We consider the photon energy ω which is much larger than the ionization potential I, i.e. $\omega \gg I$. Limiting ourselves by the condition $\omega \ll m$ we can treat the photoelectron in nonrelativistic approximation. The kinetic energy of the photoelectron is $\varepsilon = \omega - I = p^2/2m$. The electron momentum p is much larger than the characteristic momentum $\mu = (2mI)^{1/2}$ of the bound state ($p \gg \mu$). At $\omega \gg I$ the photoionization requires large momentum $\mathbf{q} = \mathbf{k} - \mathbf{p}$ to be transferred to the recoil fullerene. Here $\mathbf{k}$ is the photon momentum, and $k = |\mathbf{k}| = \omega$. One can see that $k \ll p$ if $I \ll \omega \ll m$ and thus we can put $|\mathbf{q}| = q = p$.

If the electron–photon interaction is written in the velocity form, momentum q is transferred in the initial state in ionization of s states [11,4]. Thus the photoelectron can be described by plane wave. Interaction of the photoelectron with the ionized fullerene provides the contributions of the relative order $O(1/p)$ to the amplitude. Hence they contribute to the cross section beyond the asymptotics. The photoionization amplitude can be written as $F = n(\omega)\int d^3r\psi_\mathbf{p}^*(\mathbf{r})\gamma\psi(r)$ with $\psi_\mathbf{p}$ and ψ the wave functions of the photoelectron and the bound electron correspondingly; $\gamma = -i\sqrt{\alpha}\mathbf{e}\cdot\nabla/m$ is the operator of interaction between the photon and electron. In momentum space the amplitude takes the form

$$F = \sqrt{\alpha}n(\omega)\int\frac{d^3f}{(2\pi)^3}\psi_\mathbf{p}(\mathbf{f})\frac{\mathbf{e}\cdot\mathbf{f}}{m}\psi(\mathbf{f} - \mathbf{k}).$$

Since the photoelectron is described by the plane wave, i.e. $\psi_\mathbf{p}(\mathbf{f}) = (2\pi)^3\delta(\mathbf{f} - \mathbf{p})$, the amplitude of photoionization can be written as [11]

$$F = N(\omega)\frac{\mathbf{e}\cdot\mathbf{p}}{m}\psi(p); \quad N(\omega) = \left(\frac{4\pi\alpha}{2\omega}\right)^{1/2}. \tag{7}$$

We replaced q by p in the argument of the Fourier transform of the wave function of the fullerene electron. The latter is $\psi(p) = \int d^3r\psi(r)e^{-i\mathbf{p}\cdot\mathbf{r}}$.

Now we present the wave function $\psi(p)$ in terms of the Fourier transform of the potential

$$V(p) = \int d^3rV(r)e^{-i\mathbf{p}\cdot\mathbf{r}} = \frac{4\pi}{p}\int_0^\infty drrV(r)\sin pr. \tag{8}$$

The function $\psi(p)$ can be expressed by the Lippmann–Schwinger equation [4]

$$\psi = \psi_0 + G(\varepsilon_B)V\psi, \tag{9}$$

with G the electron propagator of free motion, $\varepsilon_B = -I$ is the energy of the bound state. The matrix element of the propagator is

$$\langle\mathbf{f}_1|G(\varepsilon_B)|\mathbf{f}_2\rangle = g(\varepsilon_B, f_1)\delta(\mathbf{f}_1 - \mathbf{f}_2); \quad g(\varepsilon_B, f_1) = \frac{1}{\varepsilon_B - f_1^2/2m}.$$

For a bound state $\psi_0 = 0$, and thus Eq. (9) can be evaluated as $\psi(p) = \langle\mathbf{p}|GV|\psi\rangle = g(\varepsilon_B, p)J(p)$ with

$$J(p) = \int\frac{d^3f}{(2\pi)^3}\langle\mathbf{p}|V|\mathbf{f}\rangle\langle\mathbf{f}|\psi\rangle = \int\frac{d^3f}{(2\pi)^3}V(\mathbf{p} - \mathbf{f})\psi(\mathbf{f}). \tag{10}$$

Putting also $g(\varepsilon_B, p) = -2m/p^2$ we obtain

$$\psi(p) = -\frac{2m}{p^2}J(p) = -\frac{J(p)}{\omega}. \tag{11}$$

The integral $J(p)$ is saturated at $f \sim \mu \ll p$. Thus its dependence on p is determined by that of $V(p)$. Another presentation

$$J(p) = \int d^3r\psi(r)V(r)e^{-i\mathbf{p}\cdot\mathbf{r}} = \frac{4\pi}{p}\int_0^\infty dr\chi(r)V(r)\sin(pr);$$

$$\chi(r) = r\psi(r), \tag{12}$$

which can be obtained by the Fourier transformation of the integrand on the right-hand side of Eq. (10).

We shall demonstrate that for the potential can take the form $V(p) = V_1(p) + V_2(p)$ with the two terms corresponding to two fullerene characteristics R_1 and R_2. In this case $J(p)$ can be presented as

$$J(p) = V_1(p)\kappa_1 + V_2(p)\kappa_2, \tag{13}$$

where the factors $\kappa_{1,2}$ do not depend on p, being determined by the characteristics of the bound state. Thus Eq. (11) can be written as

$$\psi(p) = -\frac{1}{\omega}\left(V_1(p)\kappa_1 + V_2(p)\kappa_2\right), \tag{14}$$

and Eq. (7) can be presented as

$$F = -\frac{N(\omega)}{\omega}\frac{\mathbf{e}\cdot\mathbf{p}}{m}\sum_{i=1,2}V_i(p)\kappa_i. \tag{15}$$