Angle-differential elastic-electron scattering off $C_{60}$: a simple semi-empirical theory versus experiment

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Received 18 November 2018, revised 3 March 2019
Accepted for publication 14 March 2019
Published 2 April 2019

Abstract

We prove in the present paper that simple modeling of a complicated highly polarizable $C_{60}$ target by a rectangular (in a radial coordinate) square well potential in combination with the static polarization potential provides a viable approximation for a low-energy elastic-electron scattering off this target. The proof is based on the results of the comparison of the calculated angle-differential elastic-electron scattering cross section off $C_{60}$ versus corresponding experimental data.

Keywords: electron scattering, fullerene, angular distribution

(Some figures may appear in colour only in the online journal)

1. Introductory remarks

In this paper, we suggest a simple method that presents a relatively accurate first step approximation to low-energy elastic-electron scattering off a $C_{60}$ fullerene. Fullerenes and endohedrals $A@C_{60}$ are very complex multi-atomic objects. This makes $ab$ initio calculations of electron scattering cross sections by these objects a very hard, if not unachievable task, and to develop theoretical approaches to this process is a challenge.

This is why low-energy elastic-electron scattering off a $C_{60}$ or $A@C_{60}$ endohedral fullerene has been in the focus of theorists for some time now, see, e.g. [1–8] and references therein. Whereas works [1, 2] addressed the problem starting from the first $ab$ initio principles, already a part of the work [2] as well as works [4–6, 8] used a semi-empirical approach. There the $C_{60}$ cage was modeled by a rectangular (in a radial coordinate) square well spherically symmetric potential, $U_C(r)$, of certain inner radius, $r_0$, thickness, $\Delta$, and depth, $U_0$, to address a low-energy elastic $e^- + C_{60}$ scattering [2, 4–6] or $e^- + A@C_{60}$ scattering [6, 8]:

$$U_C(r) = \begin{cases} -U_0, & \text{if } r_0 \leq r \leq r_0 + \Delta, \\ 0, & \text{otherwise} \end{cases}.$$ (1)

Works [2, 4, 5] suffer a serious drawback, since they neglect even in the frame of spherically symmetric approximation the effect of polarization of $C_{60}$ upon the scattering process. This effect cannot be small since $C_{60}$ cage is a highly polarizable object. Indeed, the dipole static polarizability, $\alpha_{C_{60}}$, of $C_{60}$ is approximately 850 at. un. [9] (at. un. stands for ‘atomic units’ which is the system of units used throughout the present paper unless stated otherwise). Further corrections to the model, made in works [6, 8], improved the approximation considerably by introducing a new model potential $U_{C_0}(r)$ of a polarizable $C_{60}$ as the sum of the $U_C(r)$ potential and the static polarization potential $V_{v_0}(r)$:

$$U_{C_0}(r) = U_C(r) + V_{v_0}(r),$$ (2)

$$V_{v_0}(r) = -\frac{\alpha_{C_{60}}}{2(r^2 + b^2)^2}.$$ (3)
Here, \( b \) is the parameter of the order of the size of the target (C\(_{60}\) in our case) that prevents the unphysical divergence of the potential at \( r \to 0 \) and permits to satisfy the obvious physical condition \( V_c(r) |_{r \to 0} = \text{const} \). In the present paper, the \( b \) parameter is put to \( b \approx 8 \) which is approximately the size of C\(_{60}\).

The polarization potential \( V_p(r) \) originates from physics of atom, where it was introduced in studies of a concrete negative ion \( O^- \) with \( \alpha_C \) treated as an adjustable parameter in [10] yet in 1943. This potential was developed by introducing energy dependence of \( \alpha_C \), but has remained a phenomenological one [11]. A new approach, based on a many-body diagrammatic technique suitable for atoms, leads to good results without any adjustable parameters (see [12] and references therein). However, this approach requires much more complicated calculations even for atoms.

2. Essential formulas

Although quite accurate, the \textit{ab initio} many-body theory approach [12] is extremely difficult to apply to such a tremendously complex objects as fullerenes and endohedrals. This justifies using an approach that in principle is much less accurate, but at the same time much simpler [6]. However, in the absence of the comparison with corresponding experimental data for elastic \( e^- + C_{60} \) or \( e^- + A@C_{60} \) scattering it has remained unclear how viable are the approximations above, equations (1)–(3), when applied to the scattering process in question. Whereas we are not aware of the existence of experimental data for the integral, i.e. total elastic scattering cross section, \( \sigma_{tot} \), we have become aware of a long-time existing experimental data [13] for the angle-differential elastic scattering cross section, \( d\sigma/d\Omega \), in the energy region up to about 12 eV of the electron impact energy \( E \). It is the aim of the present paper to demonstrate the viability of the simple spherically symmetric phenomenological model, equations (1)–(3), and to demonstrate thus that reasonably accurate results can be obtained in the frame of a very simple and easily generalizable to other than C\(_{60}\) targets approach.

To meet the goal, we calculate in the present work \( d\sigma/d\Omega \) for \( e^- + C_{60} \) scattering, obtained with the utilization of equations (2) and (3), and compare the thus obtained result with experiment [13] for a number of scattering angles \( \theta = 30^\circ, 70^\circ, 80^\circ \) and \( 90^\circ \). We demonstrate a reasonably good agreement between measured and calculated data.

We substitute the real field of fullerene C\(_{60}\) that has a very complex structure and is spherically symmetric only on the average by a spherically symmetric potential. Thus, we limit the consideration by relatively low scattering energies, \( E \), at which the incoming electron wavelength is essentially bigger than the distance between two adjacent carbon atoms of the fullerenes shell. However, by neglecting the detailed structure of the C\(_{60}\) shell, we simplify the treatment of \( e^- + C_{60} \) scattering problem drastically, acquiring the possibility to apply to it the powerful apparatus developed in quantum mechanic to treat electron scattering by potential well.

The first step on this way is presentation of the incoming electron wave as a sum of partial angular momentum contributions. Then the angle-differential scattering cross section is calculated using the well-known formula for scattering by a spherically symmetric potential, see, e.g. [14]:

\[
\frac{d\sigma(E, \theta)}{d\Omega} = \frac{1}{k^2} \sum_{l, l'} (2l + 1)(2l' + 1) \sin \delta_l \sin \delta_{l'} \times \cos(\delta_l - \delta_{l'}) P_l(\cos \theta) P_{l'}(\cos \theta).
\]  

(4)

Here, and everywhere else in this paper, \( k \) is the electron momentum, \( k = \sqrt{2E} \), \( \theta \) and \( \Omega \) are the scattering angle and solid angle, respectively, \( \delta_l \) are scattering phase shifts and \( P_l(\cos \theta) \) is the Legendre polynomial of the \( l \)th order.

The total electron elastic scattering cross section, \( \sigma_{tot}(k) \), is calculated in accordance with the standard formula for electron scattering by a spherically symmetric central-potential field:

\[
\sigma_{tot}(k) = \frac{4\pi}{k^2} \sum_{l=0}^\infty (2l + 1) \sin^2 \delta_l(k).
\]

(5)

The radial parts \( R_{2l}(r) \), of the wave functions of scattering states with the definite energy \( \varepsilon = k^2/2 \), \( \Psi_{l,2l+1}(r) = (R_{2l}(r)Y_{l,2l}(\theta, \phi) \) are obtained by solving the corresponding radial Schrödinger equation.

Once \( R_{2l}(r) \) are determined, the needed electron elastic scattering phase shifts, \( \delta_l(k) \), are found by referring to \( R_{2l}(r) \) at \( r \gg 1 \):

\[
R_{2l}(r) \to \sqrt{\frac{2}{\pi}} \sin \left( kr - \frac{\pi l}{2} + \delta_l(k) \right).
\]

(7)

We determine the scattering phase shift \( \delta_l(k) \) from a numerically derived wave function \( R_{2l}(r) \) using a computing code, presented in [15].

3. Results of calculations

In the calculations, we use two different sets of the adjustable parameters \( r_0, \Delta \) and \( U_0 \) for the \( U_C \) cage potential, equation (1), that exist in literature. One of the two sets of the parameters, referred to as the ‘set 1’, is \( r_0^{(1)} = 5.8, \Delta^{(1)} = 1.9, U_0^{(1)} = 0.302 \) at. un., as in, e.g. [16] and references therein; the other one, referred to as the ‘set 2’, is \( r_0^{(2)} = 5.26, \Delta^{(2)} = 2.91, U_0^{(2)} = 0.260 \) at. un., as in, e.g. [2, 8] and references therein. The use of two sets of parameters permits to test the sensitivity of the results to parameters variation.

The utilization of the energy independent \( U_C(r) \), as well as the omission of separate electron exchange potential, simplifies the calculations drastically. Note, however, that we choose only such combination of pseudo-potential (1) parameters \( U_0 \) and \( \Delta \) that leads to a \( C_{60} \) electron affinity coinciding with experimentally measured values. Therefore, \( U_C(r) \) effectively accounts for all the short-range interaction,
including exchange. The neglect of possible energy dependence of \( U_{rC} \) is inessential since we consider low-energy scattering only.

Contrary to the \textit{ab initio} many-body approach \cite{17}, potential \( U_{rC}(r) \) permits easily calculate scattering phase shifts \( \delta_\ell \) almost irrespectively to the value of \( \ell \). So, we performed a trial calculation that showed that partial electronic waves with the orbital momentum \( \ell \) up to \( \ell = 2 \) is the sufficient number of terms to be accounted in the calculation of \( d\sigma/d\Omega \) in order to ensure the convergence of the sum in equations (4) and (5).

Corresponding calculated data for \( d\sigma/d\Omega \), obtained without account for the \( V_0 \) polarization potential, equation (3), i.e. with the use of \( U_C(r) \) determined by equation (1) alone, are depicted in figure 1, whereas those obtained with the account for the \( V_0 \) potential, i.e. with the use of \( U_{rC}(r) \), equation (2), are depicted in figure 2.

One can see, that the account for \( C_{60} \) polarization potential, figure 2, corrects \( d\sigma/d\Omega \) considerably, especially at \( \theta = 30^\circ \) and \( 70^\circ \), making the agreement between the present theory and experiment be quite reasonable. This agreement, the authors dare to state, is, in the whole, even somewhat better than that between the \textit{ab initio} theory \cite{1} and experiment. As for the comparison of results, obtained for \( d\sigma/d\Omega \) with the use of the parameter set 1 and parameter set 2 for \( r_U \), \( U_0 \) and \( \Delta \), it is difficult to give an unambiguous preference to the one over the other.

Some peculiar features deserve attention. The positions of resonances at \( 30^\circ \) in our calculations are 2 (set 1) and 3 (set 2) eV that is close to 3 eV from \cite{1}, while experiment gives 4.2 eV. At \( 70^\circ \) our calculations predict resonances at 9.3 (set 1) and 9 (set 2) eV that is close to experimental position 9 eV, both in energy and in magnitude. For such a simplified approach, the agreement for the first maximum, 3.5 eV, for both sets as compared to 4 eV in experiment, is quite acceptable. Similar is the situation for other angles. We managed also to reproduce the 7 eV resonance at \( 80^\circ \) predicted in \cite{1}, in good agreement with experimental data. Our calculations at \( 80^\circ \) reproduce the position of the resonance at 2 eV, predicted in \cite{1}, but attribute to it a much smaller width.

Figure 1. \( d\sigma/d\Omega \) (in units \( a_0^2 \text{ sr}^{-1} \), \( a_0 \) being the first Bohr radius) for \( e^- + C_{60} \) scattering at \( \theta = 30^\circ, \ 70^\circ, \ 80^\circ \) and \( 90^\circ \) as a function of \( E \). Solid and dashed lines are results of the present calculation obtained without account for polarizability of \( C_{60} \) with the use of, respectively, the set 1 and set 2 for \( r_U, \Delta \) and \( U_0 \). Solid squares, experiment \cite{13}. Note: the experimental points are copied from \cite{1} ’as is’, in order to not distort the comparison of theory \cite{1} with experiment in other parts of the present paper.
Solid and dashed squares are the experiment [13]. Solid circles are results of the ab initio theory [1]. As in figure 1, both the experimental points (solid squares) and results of the ab initio theory [1] (solid circles) have been copied from [1] ‘as is’ in order to not distort the comparison of theory [1] with experiment in any way.

In figure 3 we present results of another type of differential in angle elastic scattering cross sections. Namely, the dependences of $d\sigma(E, \theta)/d\Omega$ (in units $a_0^2 \text{s}^{-1}$, $a_0$ being the first Bohr radius) for $e^- + C_{60}$ scattering at $\theta = 30^\circ$, $70^\circ$, $80^\circ$ and $90^\circ$ as a function of $E$. Solid and dashed-dotted lines are results of the present calculation obtained with account for polarization potential of $C_{60}$ with the use of, respectively, the set 1 and set 2 for $r_0$, $\Delta$ and $U_{0}$. Solid squares are the experiment [13]. Solid circles are results of the ab initio theory [1]. As in figure 1, both the experimental points (solid squares) and results of the ab initio theory [1] (solid circles) have been copied from [1] ‘as is’ in order to not distort the comparison of theory [1] with experiment in any way.

Figure 3. $d\sigma(E, \theta)/d\Omega$ (in units $a_0^2 \text{s}^{-1}$, $a_0$ being the first Bohr radius) for $e^- + C_{60}$ scattering at $E = 0.018$, $0.038$, $0.073$, and $0.109$, all in Ry, as a function of $\theta$. Upper part of the figure presents results with polarization potential, while the lower—the same, but without polarization potential.

The usual estimation of the required upper value of the partial wave and corresponding scattering phase shifts taken into account is $\ell_{\text{max}} \approx kr_0$. In the considered case, one has $\ell_{\text{max}} \approx 6$ even for the highest considered incoming electron energy. This is well below the upper $\ell$ value 32, included in our calculations. This estimation is valid for all short-range potentials, namely those that decrease faster than $1/r^2$ with distance $r$ growth.

Note, however, that the differential in angle scattering cross sections $d\sigma(E, \theta)/d\Omega$ require much more scattering phase shifts than the total cross section $\sigma$, since the smallest phase shift $\delta_{\ell_{\text{max}}}$ that is taken into account contributes to $d\sigma(E, \theta)/d\Omega$ a term $\sim \sin \theta_{\text{max}}$, while to $\sigma$ the contribution of this same phase shift is much smaller, since the cross section $\sigma$ is proportional to $\sin^2 \theta_{\text{max}}$ and an inequality $\sin^2 \theta_{\text{max}} \ll \sin \theta_{\text{max}}$ is valid.
for $\delta_{l_{\text{max}}} \ll 1$. In general, $d\sigma(E, \theta)/d\Omega$ is much more sensitive to the details of potential employed in cross section calculations than $\sigma$. This is why agreement achieved with experimental data in $d\sigma(E, \theta)/d\Omega$ is a strong evidence of existence of even better agreement for $\sigma_{\text{tot}}(k)$ values.

To illustrate the role of electron partial waves with high angular momentum and corresponding low scattering phase shifts in forming $d\sigma(E, \theta)/d\Omega$, we have calculated this differential cross section with first six scattering phase shifts ($l = 0 - 5$), and total set of the considered $l = 0 - l_{\text{max}}$ ($l$ - All) with $l_{\text{max}} = 32$, for a number of energies, $E = 0.09, 0.49, 1.00$, all in Ry.

The results are presented in figure 5. The upper part depicts $d\sigma(E, \theta)/d\Omega$ calculated for these two sets of $l$. While for $E = 0.09$ Ry curves for $l = 0 - 5$ and $l = 0 - l_{\text{max}}$ are almost indistinguishable, the situation for $E = 0.49$ Ry is already completely different, since scattering phase shifts $l > 5$ contribute a lot, particularly at small scattering angles. For $E = 1.00$ Ry the contribution of higher than $l > 5$ waves becomes decisively important.

The lower part of figure 5 presents $d\sigma(E, \theta)/d\Omega$ that we have calculated taking into account the contribution of all $l \leq 32$ scattering phase shifts for electron energies $E = 0.09, 0.49$ and 1.00, all in Ry. We see the general tendency: with energy growth differential in angle scattering cross section $d\sigma(E, \theta)/d\Omega$ becomes more and more anisotropic and the number of oscillations in it increases.

Presenting concrete results of calculations for $d\sigma(E, \theta)/d\Omega$ dependences upon both variables has as a primary aim the demonstration of simplicity, flexibility, and effectiveness on the proposed method.

4. Conclusions and discussions

In conclusion, the provided in the present paper results prove the applicability of the $U_{C_60}$ potential (2) to tackling the quite hard problems of electron scattering off a highly polarizable $C_{60}$ fullerene in a rather simple way. Note that, obviously, by a fine-tuning of the parameters $r_0$, $U_0$ and $\Delta$, as well the polarization parameters $\alpha$ and $b$, one can achieve a yet better agreement between theory and experiment than the present one. As to the number of scattering phase shifts that is necessary to take into account, in the frame of the spherically symmetric approximation it is for a given potential (1–3) totally determined by the incoming electron energy. One would have been too naïve, however, to have expected more from the utilized simple modeling of $C_{60}$ and yet its surprising viability is obvious, at least in the aim of getting the insight into the polarization potential effects in electron scattering of $C_{60}$.

Our aim was different from that in [1–3]. Instead of achieving the best possible theoretical description of the $e + C_{60}$ process regardless to efforts required, we had in mind to present a method that while being simple enough could guarantee a reasonable agreement of calculation results with general features of experimental data. The obvious advantage of our method is its ability to consider easily other than $C_{60}$ fullerenes using available computing codes.
The suggested approach permits to obtain results relatively easy, thus opening up an option to obtain good and relatively reliable pre-experimental data in course of preparation of accurate very hard measurements. We do not expect, however, that such a crude approach could present reliable predictions of tiny features of experimental data, such as e.g. narrow resonances.

Note, that in a manner, similar to developed in this paper, the potentials (2, 3) one could generalize to treat also endohedrals $A@C_{60}$ as targets in $e + A@C_{60}$ or even in $e + A@C_N$ elastic scattering processes.

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**References**

[1] Gianturco F A, Lucchese R R and Sanna N 1999 *J. Phys. B: At. Mol. Opt. Phys.* **32** 2182
[2] Winstead C and McKoy V 2006 *Phys. Rev. A* **73** 012711
[3] Felfli Z and Msezane A Z 2018 *Eur. Phys. J. D* **72** 78
[4] Dolmatov V K, Cooper M B and Hunter M E 2014 *J. Phys. B: At. Mol. Opt. Phys.* **47** 115002
[5] Dolmatov V K, Bayens C, Cooper M B and Hunter M E 2015 *Phys. Rev. A* **91** 062703
[6] Amusia M Y and Chernysheva L V 2015 *JETP Lett.* **101** 503
[7] Amusia M Y and Chernysheva L V 2016 *JETP Lett.* **103** 286
[8] Dolmatov V K, Amusia M Y and Chernysheva L V 2017 *Phys. Rev. A* **95** 012709
[9] Amusia M Y and Baltenkov A S 2006 *Phys. Lett. A* **360** 294
[10] Bates D R and Massey H S 1943 *Trans. R. Soc. A* **239** 269
[11] Drukarev G F 1987 *Collisions of Electrons with Atoms and Molecules, Physics of Atoms and Molecules* (Berlin: Springer)
[12] Amusia M Y, Chernysheva L V and Yarzhemsky V G 2012 *Handbook of Theoretical Atomic Physics, Data for Photon Absorption, Electron Scattering, and Vacancies Decay* (Berlin: Springer) p 812
[13] Tanaka H, Boesten L, Onda K and Ohashi O 1994 *J. Phys. Soc. Japan* **63** 485
[14] Landau L D and Lifshitz E M 1981 *Quantum mechanics, Non-Relativistic Theory* 3rd edn (Oxford: Butterworth-Heinemann)
[15] Amusia M Y and Chernysheva L V 1997 *Computation of Atomic Processes: A Handbook for the ATOM Programs* (Bristol: Institute of Physics)
[16] Connerade J P, Dolmatov V K and Manson S T 1999 *J. Phys. B: At. Mol. Opt. Phys.* **32** L395
[17] Amusia M Ya and Cherepkov N A 1975 Many-electron correlations in the scattering processes *Case Studies Atom. Phys.* **5** 47

**Figure 5.** $d\sigma(E, \theta)/d\Omega$ (in units $a_0^2 sr^{-1}$, $a_0$ being the first Bohr radius) for $e^- + C_{60}$ scattering at $E = 0.09, 0.049,$ and $1.00$, all in Ry, as a function of $\theta$. Upper part of the figure compares results for $l = 0 - 5$ and $l = 0 - l_{\text{max}}$, while the lower presents results for $l = 0 - l_{\text{max}}$ only.