Elastic scattering of positrons by H_2O at low energies

J.L.S. Lino
Assessoria e Orientação Estudantil - AOE,
Cep:12243-010, n° 80São José dos Campos, São Paulo, Brasil

Received 19 June 2013; accepted 31 January 2014

We present a study theoretical on elastic positron-H_2O collisions. More specifically, integral, and differential cross sections in the 1-10 eV energy range are reported. The calculations were performed using the Schwinger multichannel method with plane waves as a trial basis set (Jorge L S Lino, Phys. Scr. 80 (2009) 065303). The positron-H_2O interaction is described using the static plus polarization contributions. Comparison of our calculated results with the recent experimental of Zecca *et al* (J.Phys.B 39 (2006) 1597) and theoretical results of Arretche *et al* (Nucl. Instr. Meth. Phys. B 268 (2010) 178) is encouraging.

Keywords: Schwinger; pósitron; water.

Se analiza uma aplicacion del principio variacional de Schwinger desde la perspectiva de ondas planas para um conjunto base (Jorge L S Lino, Phys. Scr. 80 (2009) 065303). y seccion integral and eficaz diferencial para positron- H_2O en el intervalo de 1-10 eV. Los resultados obtenidos se comparam con los experimentos and (Zecca *et al* (J. Phys.B 39 (2006) 1597) y teoricos (Arretche et al Nucl. Instr. Meth. Phys. B 268 (2010) 178).

Descriptores: Schwinger; positrón; agua.

PACS: 36.10.Dr; 34.50.Gb

1. Introduction

Positron-matter interactions play an important role in an increasingly broad variety of applications, for example, positrons injected into matter thermalize quite rapidly and, in many insulating compounds, some of the positrons form various states of positronium [1,2]. The thermalised positron can interact with the molecular medium and give rise to a large variety of energy transfer processes similar to those occurring with electrons as projectiles. The physics involved in positron scattering processes can be useful for understanding practical systems as, for example, in positron annihilation in solids [3], scans of the human brain in high-resolution positron emission tomography [3], and in the spectrum analysis of emission of radiation by positron annihilation, which can provide important information about a number of subjects of astrophysical interest, such as the nature of matter of the interstellar medium (see Refs. 3, 4). A series of experiments involving positron impact with molecules has been done recently [4] so that the theoretical calculations are extremely important for an adequate comparison. Actually the atomic and molecular physics community is working to understand the dynamics of positron with the target environments, *i.e.*, direct annihilation and formation processes. The comprehension of these elastic and inelastic processes may be very useful in the near future to create alternative techniques to produce positrons beams (to cite positron-molecule collisions is a controversial area and several considerations have been discussed [5-12]. As controversial, for example, the influence of positronium formation on elastic positron-molecule collision is still on open problem which deserves both theoretical and experimental efforts, second, the possible origin of the observed structures (positronium), present to different extents in most measurements, remains the subject of some speculations [5] and in the low energy region, early measurements in the limit zero energy on targets as N_2 indicated positron cross sections rising with decreasing energy, but more recent experiments indicate the contrary [5]). The interest of theoretical physicists has therefore shifted considerably from atoms, molecules and larger molecular targets. However, in contrast to electronic structure calculations, there are no universal tools, such as standard basis sets, that could be applied to different molecules. A suitable theoretical method should be capable of dealing with several important aspects of the problem such as polarization effects, resonances, electronically inelastic scattering with several open and closed channels, positronium formation and, if possible, with computer codes applicable to targets in general. For the low-energy regime, below all electronic and positronium formation thresholds the theoretical task is also difficult since it depends on a good description of polarization effects. The polarization potential is always attractive and somehow is always trying to put positrons and electrons together. The description of the polarization interaction is very critical because the static potential is of opposite sign from the polarization potential (the net potential is weaker and more sensitive than that of the electron case). As a result the positron-molecule problem is indeed more difficult to solve than that of the electron case, making the theoretical task of describing the phenomena very challenging. There are limited investigations, both theoretical and experimental, on positron collision with water molecules, particularly in the low impact energy. Water is one of the most important substances that compose all living organisms. For instance, about 70 % of human body, in weight, is formed by this component. In this sense, it is clear that knowledge on the interaction of all kind of radiations, mainly ionizing radi-

ations, with water is imperative (the idea of using positrons as a tool for scanning the human body in order to identify tumors). Recently Champion and Le Loirec [13] developed a code to simulate the positron path until its annihilation in a water medium, which is a common strategy to simulate biological matter.

On the theoretical side, the literature for positron scattering by water is scarce. Baluja and Jain [14] reported integral cross sections (ICS) for elastic and inelastic positron-H_2O scattering in the (10-500 eV) using a spherical symetrical optical potential approach. More recently, Baluja *et al* [15] reported rotational elastic and excitation cross sections for positron-H_2O scattering calculated using R-matrix approach. In a recent work, Arretche *et al* [16] reported elastic positron-H_2O collisions in the 0-10 eV energy range using two methods, the Schwinger multichannel (SMC) method and the method of continued fractions (MCF) [16,17]. Studies using different methods are very interesting and would allow a more critical interpretation of the agreements and discrepancies. In general, Arretche *et al* [16] obtained good agreement between the calculated results using these two methods but some differences in the shape of differential cross sections are observed. Here, we present a study of the Schwinger multichannel (SMC) method with plane waves as a trial basis set (SMC-PW) for positron-H_2O scattering. In particular, the main limitation of the SMC method established by Germano and Lima [18] resisdes on what makes it a general method: the expansion of the scattering functions is very effective for short range potentials.

Then we suggested that, for a given trial scattering wave function, the new functional should give a more adequate cross section than the original SMC method. A important development of the SMC method would be to allow inclusion of plane waves in the scattering basis set and we have developed computer codes involving matrix elements of the second Born terms with no restrictions on the molecular geometries. For example, for singlet transitions, the long-range character of the potential requires the use of a large number of partial waves to properly describe the scattering in forward direction. To define a good Cartesian Gaussian-CG basis set to produce a scattering calculation is not an easy task and the experience gained in *ab initio* calculations for positron-molecule collisions can be useful, there are no clear rules or procedures to construct trial scattering basis sets for *ab initio* scattering and a serious difficulty with CG are identified for positron-molecule (for example, instability related to the choice of the variational basis set and spurious results. One such specific area with problem is the excitation of electronic states of molecules by positron impact where studies on the electronic excitation of molecules are limited and there are reported results only for H_2 and N_2 (excitation of molecules remains controversial and the dynamical features of the annihilation process are not fully understood). Motivated by these questions we have taken up the task to investigate the SMC-PW method. In a recent work, we have reported elastic and inelastic cross sections for positron-H_2 [19] using the SMC-PW compared with the SMC traditional [18] and the close coupling approximation (CCA) [20]. In this work we report calculated differential and integral cross sections (DCS) for elastic scattering of positrons by H_2O in the low-energy range (1- 10 eV). The organization of this paper is as follows. In Sec. 2 we describe the theoretical method used; in Secs. 3, and 4 we present our numerical developments and in Sec. 5 we present results and discussions.

2. Formalism

The Hamiltonian for the (N+1)-particle collision system can be written as

$$\mathrm{H} = (\mathrm{H_N} + \mathrm{T_{(N+1)}}) + \mathrm{V} = \mathrm{H}_o + \mathrm{V} \tag{1}$$

where $\mathrm{H_N}$ is the target Hamiltonian, $\mathrm{T_{(N+1)}}$ is the kinetic energy operator for the incident particle (N is the number of the electrons of the target and + 1 is the particle incident), and V is the interaction between the scattered positron and the nuclei and electrons of the target. Here $|\mathrm{S}_{ki}\rangle$ is the input channel state represented by the product of a plane wave $\mathbf{k}_i$ times $|\Phi_o\rangle$, the initial (ground) target state. $|\mathrm{S}_{kf}\rangle$ has an analogous definition, except that the plane wave points to $\mathbf{k}_f$. The complete scattering wave function can be shown to satisfy a modified Lippmann-Schwinger equation

$$\mathrm{A}^{(+)}|\Psi^{(+)}_{\mathbf{kf}}\rangle = \mathrm{V}|\mathrm{S}_{\mathbf{kf}}\rangle \tag{2}$$

$$\mathrm{A}^{(+)} = \mathrm{V} - \mathrm{V}\,\mathrm{G}^{(+)}_{\mathbf{p}}\mathrm{V} \tag{3}$$

and $|\mathrm{S}_{\mathbf{kf}}\rangle$ are solutions of the unperturbed Hamiltonian Ho, and $\mathrm{Gp}^{(+)}$ is the projected outgoing-wave Green's function [18]. Based on the inhomogeneous Eq. (2), the bilinear variational form of the scattering amplitude is given by

$$f(\mathbf{k}_f, \mathbf{k}_i) = -(1/2\pi)\{\langle \mathrm{S}_{\mathbf{kf}}|V|\Psi^{(+)}\rangle + \langle\Psi^{(-)}|\mathrm{V}|\mathrm{S}_{\mathbf{ki}}\rangle - \langle\Psi^{(-)}|\mathrm{V\text{-}VG}_o\mathrm{V}|\Psi^{(+)}\rangle\} \tag{4}$$

The scattering states $|\Psi^{(+)}_{\mathbf{ki}}\rangle$ and $\langle\Psi^{(-)}_{\mathbf{kf}}|$ are products of the target wave function $|\Phi_\ell\rangle$ and one-particle scattering wave function. The initial step in our calculations is to expand the one-particle scattering wave functions as a linear combination of plane waves. So, the expansion of the scattering wave function is written in a discrete form as

$$|\Psi^{(+)}_{\mathbf{ki}}\rangle = \sum a_m(\mathbf{k}_\mathrm{m})|\Phi_\ell \mathrm{k}_m\rangle$$

$$|\Psi^{(-)}_{\mathbf{kf}}\rangle = \sum b_n(\mathbf{k}_n)|\Phi_\ell \mathrm{k}_n\rangle \tag{5}$$

Use of Eq. (4) and application of a stationarity condition with respect to the expansion coefficients, gives the working form of the scattering amplitude

$$f(\mathbf{k}_f, \mathbf{k}_i) = -(1/2\pi)\{\langle \mathrm{S}_{\mathbf{kf}}|\mathrm{V}|\Phi_\ell \mathrm{k}_\mathrm{m}\rangle x (\mathrm{d}^{-1})_{\mathrm{mn}}\langle \mathrm{k_n}\Phi_\ell|\mathrm{V}|\mathrm{S_{ki}}\rangle$$

where

$$d_{mn} = \langle k_n \Phi_\ell | V - V G_p^{(+)} V | \Phi k_m \rangle. \quad (6)$$

Here $G_p^{(+)}$ is the projected outgoing-wave Green's function, and $P$is the target-space unit operator,

$$G_p^{(+)} = P G_o^{(+)}$$

$$P = \sum\!\!\!\!\!\!\int |\Phi_\ell\rangle\langle\Phi_\ell| = 1$$

where

$$G_o^{(+)} = \lim_{n\to\infty} (E - H_0 + in)^{-1} \quad (7)$$

is the interaction-free Green's function and Ho is the interaction-free Hamiltonian and P is truncated and carries only energetically open bound state channels as defined for the SMC method original. It should be noted that every required matrix element involves the potential V; therefore, if V is short-ranged, $\Psi_{ki}^{(+)}$ and $\Psi_{kf}^{(-)}$ need be describe only over a finite volume.

3. The $\langle S_{kf}|$ V $G_p^{(+)}$V $|S_{ki}\rangle$ term

The numerical calculation of the matrix elements from $VG^{(+)}V$ represent the most time consuming step in the SMC-PW code and requires almost the entire computational time of the scattering calculation. The $VG^{(+)}V$ term used is an old subject of substantial importance and interest, and has drawn attention of the experts in the field [21]. Our SMC-PW can be conveniently reduced to a form involving the second Born approximation, *i.e.*, for the weak interaction limit, $V \approx 0$, and scattering amplitude are obtained from first Born approximation (FBA) and valid for high-energy static calculation (in situations where the target wave functions can be considered frozen). The scattering amplitude as showed in Eq. (6) also can be rewritten as

$$[f(\mathbf{k}_f, \mathbf{k}_i)]_{\text{SMC - PW}} = \frac{(fB1)\cdot(fB1)}{(fB1) - (fB2)} \quad (8)$$

and as cited before, when V≈0 we have

$$[f(\mathbf{k}_f, \mathbf{k}_i)]_{\text{SMC - PW}} = \frac{(fB1)}{(1 - fB2/fB1)}$$

$$\approx fB1 + fB2 + \ldots = \text{Born series}$$

Hence, neglecting the second order term we may rewrite the scattering amplitude as

$$[f(\mathbf{k}_f; \mathbf{k}_i)]_{\text{SMC - PW}} \approx [f(\mathbf{k}_f; \mathbf{k}_i)]_{\text{FBA}} : \quad (9)$$

This result shows that the SMC-PW code is just an adaptation of the FBA (or SBA) and provides a way to represent cross section for high-energies (this is a important consideration). It is believed that in the Born series on the projectile-target interaction, only the two lowest terms, the so-called first and second Born terms contribute significantly. The reduction of the SMC-PW amplitude to the f_{B1} + f_{B2} approximation is an important consideration and must be adequate for rearrangement studies. It is also assumed that for the purpose of examination of the numerical quadrature, it is justifiable to calculate the second Born terms in the static (st) approximation. Hence, the task is to the optimum quadrature of the VGV term used in the SMC-PW [19]. The original strategy used by Lima and co-workers to compute the $VG^{(+)}V$ term used in the SMC method was use the spectral decomposition of a plane wave onto a finite Gaussian basis, which provides analytical expressions. Because the required number of Gaussian functions to obtain convergence increased substantially with the target's size and with the number of considered collision channels, a second method was developed. This second method was used in our formalism (SMC-PW) and is called 3dk insertion and represent the integration over linear momentum variables arising in $VG^{(+)}V$ term and are performed by numerical quadrature. In our implementation we use two different quadratures for $|\mathbf{k}_i\rangle$ and $|\mathbf{k}_f\rangle$ to avoid situations where $|\mathbf{k}_f\rangle$ and $|\mathbf{k}_i\rangle$ are too | small. The evaluation of the $|\mathbf{k}_i\rangle$ (or $|\mathbf{k}_f\rangle$) is generally associated with double quadrature mesh (θ_i, Φ_i) and (θ_f, Φ_f). The range of momentum integration $VG_p^{(+)}V$ term covers the entire phase space (for example, 0 to 1), so one encounters and infinite integral. The $VGp^{(+)}V$ nature of the problem guarantes that the integral decays rapidly to zero as the momentum variables tend to infinity. It is often useful from a practical point of view to transform the infinite integral to a finite integral. As the usual Gauss-Legendre quadrature points often used to discretize the integrals are given between 0 and 1 or between -1 and + 1, it is convenient to transform the infinite integral to a finite integral. The evaluation of the principal-value integral over [0,1) can be performed quite accurately by splitting the range into intervals [0, k_{max}] and [k_{max}, 1) with a trivial transformation for maps $0 < k < 1$ into $0 < x < 1$. These mappings imply a maximum value of k_{max} which is efficiently controlled [19,21]. To test the numerical stability of the Green's function, we have performed cross sections for some numerical quadrature (obviously, each molecule requires a special treatment). The calculations using SMC-PW were performed recently for positron-He, Ne, and positron-H_2 [22]. As expected, a higher number of radial points are needed when a high variation of charge density can be expected along the radial coordinate, as the molecule CH_4 (it can also be concluded that the number of radial points increases with the size of the molecule). The most prominent effect of the second Born approximation (SBA) is the dependence of the particle-impact (electron or positron). The first Born amplitude is proportional to the projectile charge Z, the second Born amplitude is proportional to Z^2, and their cross-product term in the cross section is proportional to Z^3, *i.e.*, changes its sign when the charge of the projectile is reverted.

4. Polarization potential

For a realistic description of low-energy positron-molecule interaction, polarization effects must be taken into account. SMC method as originally proposed by Germano and Lima [18] describes target polarization through single virtual excitations of the target wavefunction, explicitly considered in the expansion of the scattering wave function. From a formal point of view, this procedure makes the inclusion of polarization effects not practical for molecular targets with tens of electrons. This consideration suggests a procedure alternative to describe polarization effects in our formalism SMC-PW. The polarization space was then constructed following Salvat [23]

$$\mathrm{V_{pol}}(r) = -\alpha/[r^2 + r_c^2]^2 \qquad (10)$$

where "α" is the dipole polarizability of the atom (or molecule) and "r_c" represents an phenomenological cutoff parameter, which serves to prevent the polarization from diverging at r = 0 [23]. At short range the correlation terms take into account the many-body effects, whereas at long range it simulates the polarization effects. This semiempirical form of the polarization potential presents a correct asymptotic behavior. We incorporate polarization effects simply via the Born approximation and the scattering amplitude is now formed by two parts, namely:

$$f^{\mathrm{Born}} = f^{\mathrm{Born-st}} + f^{\mathrm{Born-pol}} \qquad (11)$$

where Born-st is the static part of the Born scattering amplitude Born-pol is the polarization part of the Born scattering amplitude and, in the body frame, is calculated as follows:

$$f_{\mathrm{Born-pol}} = -(2/\Delta^2) \int e^{i\Delta\cdot r} \mathrm{V_{pol}}(r) dr \qquad (12)$$

where Δ is the elastic momentum transfer vector.

5. Results

The basis used in our study was the same used in Ref. 16 and also used in the SMC and MCF method. This basis set gives a ground state energy of -76.05 a.u. and a dipole moment of 1.98 Debyes.

In Fig. 1 we compare our integral cross sections (ICS) for elastic positron-water scattering using SMC-PW, SMC [16] and MCF [16] with existing experimental data [24,25].

At such low energies, most electronic excitation channels are closet, therefore the present comparison is meaningful. The results obtained using the SMC-PW, SMC method and also MCF method agree very well with each other in the entire energy range covered herein. Quantitatively, the existing data are discrepant from each other and our SMC-PW agree better with data reported by Zecca *et al*. [26] (reasons for the discrepant between the experimental data are cited in Ref. 16).

In Fig. 2 we shows differential cross sections (DCS) at 5 eV using SMC-PW. The DCS obtained using the R-matrix method [15], SMC [16] method and MCF [16] are also show for comparison.

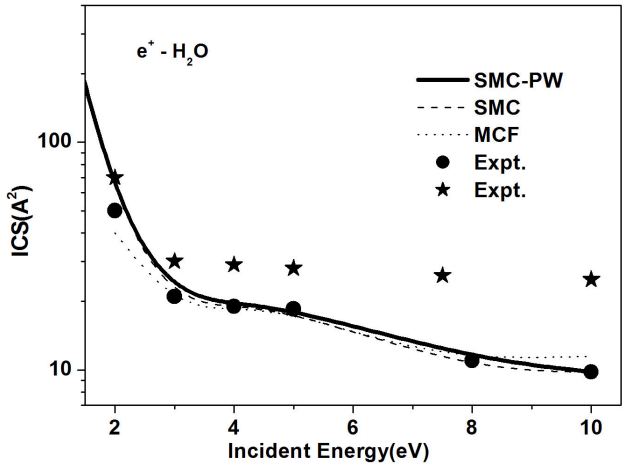

FIGURE 1. Integral cross sections (ICS) for positron-H_2O. solid line, SMC-PW method; dashed line, SMC method [16] ; dott line, MCF method [16]; circle, experimental data [24]; star, experimental data [25].

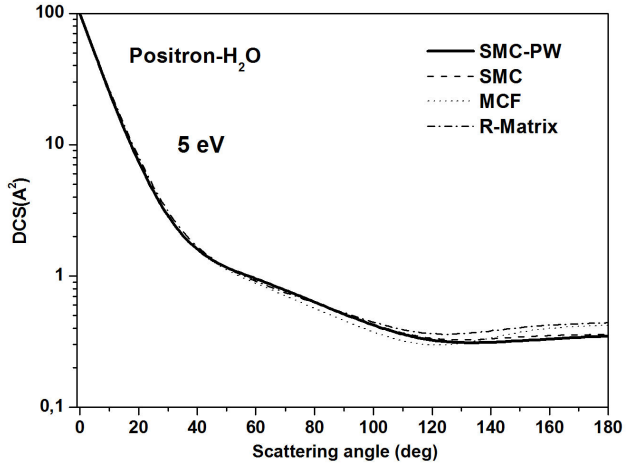

FIGURE 2. Differential cross sections (DCS) for positron-H_2O at 5 eV. solid line, SMC-PW method; dashed line, SMC method [16]; dott line, MCF method [16]; dashed long line, R-matrix, see Ref. 16.

In general, there is a good agreement between our results and theoretical studies, particularly at small scattering angles. In Figs. 3, we shows DCS at 7 eV compared with SMC method only [16]. As observed at 5 eV and 7 eV, there is a good agreement between our SMC-PW and theoretical studies, particularly at small scattering angles. Some discrepancies in shape between the DCS calculated using SMC-PW and MCF are seen and we believe that this differences are attributed due to the different manner to account for the polarization effects.

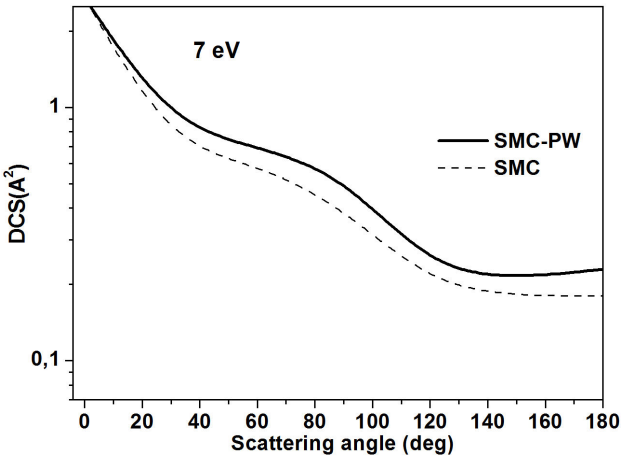

FIGURE 3. Differential cross sections (DCS) for positron-H_2O at 7 eV. solid line, SMC-PW method; dashed line, SMC method [16].

6. Conclusions

We have carried out calculations of the elastic cross sections of positron-H_2O scattering using the Schwinger multichannel method with planes wave are used as a trial basis set. Elastic integral and differential cross sections were obtained and our results are compared with the SMC and MCF methods. All calculations were performed in the fixed-nuclei approximation and part of the discrepancies between theory and experimental data can be attributed to the well known inclusion of rotational motion in the calculation and this may reduce significantly the influence of the dipole interaction. The present study suggests that the SMC-PW can be effective in the study of positron-target polar.

Acknowledgements

The author thank the Referee for fruitful discussion.

1. D. J. Day, G. Larrichia, and M. Charlton, *Hyperfine Interact.* **73** (1992) 2017.
2. T. J. Murphy, and C. M. Surko, *Phys. Rev. Lett* **67** (1991) 2954.
3. S. J. Gilbert, J. P. Sulivan, R. Greaves and C. M. Surko, *Nucl. Instr. Methods B* **171** (1992) 81.
4. G.F. Gribakin, J.A. Young and C.M. Surko, *Rev. Mod. Phys.* **82** (2010) 2557
5. T. L. Gibson, *J. Phys. B* **25** (1992) 1321.
6. T. J. Murphy, and C. M. Surko, *Phys. Rev. A* **46** (1992) 5695.
7. D. D. Reid, and J. M. Wadehra, *J. Phy. B* **29** (1996) L127.
8. J. Beale, S. Armitage, and G. Laricchia, *J. Phys. B* **39** (2006) 1337.
9. F. Arretche, and M. A. P. Lima, *Phys. Rev. A* **74** (2006) 042713.
10. E. P. da Silva, M. T. D. Varella, and M. A. P. Lima, *Phys. Rev. A* **72** (2005) 062715.
11. J. L. S. Lino, J. S. E. Germano, and M. A. P. Lima, *J. Phys. B* **27** (1994) 1881.
12. J. L. S. Lino, *Chin. J. Phys.* **44** (2006) 497.
13. C. Champion and C. Le Loirec, *Phys. Nucleonics* **11** (1953) 40.
14. A. Jain, *Phys. Rev. A* **41** (1990) 2437.
15. K. L. Baluja and A. Jain, *Phys. Rev.* **45** (1992) 7838.
16. F. Arretche *et al*., *Nucl. Instr. And Meth. B* **268** (2010) 178.
17. M. T. Lee, I. Iga, M. M. Fujimoto, and O. Lara, *J. Phys. B* **28** (1995) 3325.
18. J. S. Germano and M. A. P. Lima, *Phys. Rev. A* **47** (1993) 3976.
19. J. L. S. Lino, *Phys. Scr.* **76** (2007) 521.
20. N. J. Mason and W. R. Newell, *J. Phys. B* **20** (1987) 3913.
21. M. A. P. Lima, *et. al*., *Phys. Rev. A* **41** (1990) 327.
22. J. L. S. Lino, B. Christ, and Chin, *J. Phys.* **50** (2012) 398.
23. F. Salvat, *Phys. Rev. A* **68** (2003) 012708.
24. M. Kimura, O. Sueoka, A. Amada, and Y. Itikawa, *Adv. Chem. Phys.* **111** (2000) 537.
25. A. Zecca , D. Sanyal, M. Chakrabarti, and M. J. Brunger, *J. Phys. B.* **39** (2006) 1597.