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Positron total cross sections for collisions with O_2, H_2O and CH_4 in energy range from 30 to 3000eV*

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Total (elastic plus inelastic) cross sections (TCSs) for the scattering of positrons by molecules O_2, H_2O and CH_4 in the energy range from 30 to 3000eV are calculated using the additivity rule model at Hartree–Fock level. A complex optical model potential modified by incorporating the concept of bonded atom which takes into consideration the overlapping effect of electron clouds between two atoms in a molecule is firstly employed for the TCS calculation of positron–molecule scattering. TCSs are quantitatively compared with those obtained in experiments and other theories wherever available, and good agreement is obtained in the energy range from 30 to 3000eV. It is shown that the additivity rule model together with the modified complex optical model potential is completely suitable for the TCS calculations of the positron–molecule scattering.

Keywords: positron scattering, TCSs, additivity rule, atomic and molecular collision
PACC: 3485, 3485B

1. Introduction

It is well known that positron–molecule collision processes play an important role in numerous areas, such as gaseous lasers, industrial production, chemical physics and planetary atmosphere. Positrons, on the other hand, are very short-lived when produced in our world of ordinary matter because of their natural tendency to annihilate with electrons. During the past decades, experimental investigations of positron scattering by molecules not only provided some direct information leading to a better understanding of the above-mentioned processes, but also lead to a better understanding of approximations used in various scattering theories.[1] Positrons differ from electrons only by the sign of their electric charges. As a result, one significant difference is that the projectile electron is indistinguishable from the atomic electrons, which gives rise to an exchange interaction that is not present in positron–molecule scattering.

Based on the properties of positrons and the importance of positron–molecule scattering processes mentioned above, many experimental investigations of positron–molecule scattering have been made. Positron–molecule scattering presents a more complex problem than the corresponding positron–atom scattering due to the multi-centre nature and its nuclear motion. Thus, few accurate theoretical calculations have been carried out so far. Though many theoretical calculation methods have been proposed in the past decades,[2–5] in the intermediate- and high-energy range, these methods cannot give accurate results of total cross sections (TCSs) since acquiring a molecular wavefunction is very difficult, as almost all inelastic channels (rotational, vibrational, electronic excitation and ionization, etc) are open. It is therefore unsurprising that many previous calculations on TCSs for positron–molecule scattering are restricted to low-energy region.

Recently, a fairly simple approach, namely the additivity rule (AR) model, wherein a molecular cross section is an incoherent sum of the cross sections of the constituent atoms, was employed successfully by Raj[6] to obtain the TCSs for positron scattering by a

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sample of seven molecules (O_2, CO, CO_2, SO_2, CS_2, OCS and SF_6) at 100–500eV. Very recently, Reid and Wadehra[7] also calculate the TCSs for positron scattering from molecules consisting of C, N and O atoms at 100–5000eV. Though many authors[6,7] have given the TCSs for positron scattering by molecules through the use of the AR model, large differences are noted between calculated results and experimental data below 1000eV.

In this paper, the complex optical model potential which takes into consideration the overlapping effect of electron clouds between two atoms in a molecule is firstly employed to calculate the TCSs for positron scattering from the molecules O_2, H_2O and CH_4 at intermediate and high energies. In the next section, we describe in detail the modified potential and the AR model. In Section 3, we present the calculation results and discussion for the TCSs of the target molecules and compare them with the available results in experiments and other theories. Concluding remarks are made in Section 4. Atomic units are used throughout this paper unless otherwise specified.

2. Theory and method

TCSs $Q_{\rm T}(E)$ of the AR model for a molecule containing N atoms is given by

$$\begin{aligned} Q_{\rm T}(E) =& \frac{4\pi}{k}{\rm Im}F_{\rm m}(\theta=0) \\ \approx& \frac{4\pi}{k}{\rm Im}\sum_{j=1}^{N} f_j(\theta=0) = \sum_{j=1}^{N} q_{\rm T}^j(E), \end{aligned} \quad (1)$$

where $F_{\rm m}(\theta=0)$ is the positron–molecule scattering amplitude in the forward direction; $q_{\rm T}^j(E)$ and $f_j(\theta=0)$ are the TCS due to the jth atom of the molecule and the complex scattering amplitude for the jth constituent atom of the molecule, respectively. Here, it is obvious that no molecular geometry is involved in the AR model. So, the positron–molecule scattering problem is reduced to the positron–atom scattering problem which is easier to tackle.

In the present study, we employ the complex optical model potential method to obtain the TCS for positron scattering from the constituent atoms of the molecule. In the method, the potential of a constituent atom in the molecule can be expressed as follows:

$$V_{\rm opt}(r) = V_{\rm rel}^{+} + {\rm i}V_{\rm abs}^{+}(r), \quad (2)$$

here,

$$V_{\rm rel}^{+} = V_{\rm s}^{+} + V_{\rm p}^{+}.$$

Thus $V_{\rm opt}$ incorporates all the important physical effects. In detail, the repulsive static potential $V_{\rm s}^{+}(r)$ for an positron–atom scattering system is calculated by using the atomic charge density, determined from the well-known Hartree–Fock atomic wavefunction.[8] The parameter-free polarization potential $V_{\rm p}^{+}(r)$ was given by our group.[9] It may be pointed out that the polarization potential for positron scattering will be the same as for the electron while the repulsive static potential will have the opposite sign. And the absorption potential $V_{\rm abs}^{+}(r)$[10] represents the combined effect of all the inelastic channels, and is of the following form:

$$V_{\rm abs}^{+} = -\frac{1}{2}\rho(r)\overline{\sigma}_{\rm b}(k_{\rm F},k)\upsilon, \quad (3)$$

here,

$$\begin{aligned} \overline{\sigma}_{\rm b}(k_{\rm F},k) =& \frac{32\pi^2}{15k^2}N_{\rm k}H(k^2-w) \\ & \times\left\{\frac{5k_{\rm F}^3}{w} - \frac{5k_{\rm F}^3}{k^2} - \frac{2k_{\rm F}^5}{k^4} + H(k_{\rm F}^2+w-k^2)\right. \\ & \left.\times\left(\frac{2(k_{\rm F}^2+w-k^2)^{2.5}}{k^4}\right)\right\}, \end{aligned}$$

$$N_{\rm k} = \frac{3}{4k_{\rm F}^2\pi},$$

$$k_{\rm F} = [3\pi^2\rho(r)]^{1/3},$$

$$w = 2\Delta.$$

Here $\rho(r)$ is the atomic charge density, and Δ is the mean excitation energy. k and υ are the momentum and the local velocity of incident positron, respectively. $H(x)$ is a Heaviside function defined by $H(x)=1$ for $x \geq 0$, and $H(x)=0$ for $x<0$.

Now, in Eq.(2), we take into consideration the formation of the chemical bond and the overlapping effect of electron clouds between two atoms in a molecule. That is to say, in Eq.(2), we incorporate the concept of bonded atom.[11] According to the concept of bonded atom, electron charge density $\rho(r)$ may be modified as follows:

$$\rho'(r) = f\cdot\rho(r), \quad (4)$$

here,

$$f = 1 - \frac{r}{R}\times\frac{Z}{N},$$

where f is a structural factor; r and R are the bond length and the sum of the radius of the two atoms, respectively; Z and N are the number of electron in the outmost shell and the total number of electron in the atom, respectively. So, Eq.(2) can be modified as

$$V_{\rm opt}(r) = V_{\rm rel}^{+} + {\rm i}f\cdot V_{\rm abs}^{+}(r), \quad (5)$$

here,

$$V_{\rm rel}^{+} = f \cdot V_{\rm s}^{+} + V_{\rm p}^{+}.$$

The $q_{\rm T}^{j}$ in Eq.(1) is obtained by the method of partial waves,

$$\begin{aligned} q_{\rm T}^{j}(E) =& q_e^j(E) + q_a^j(E) \\ =& \frac{\pi}{k^2} \cdot \sum_{l=0}^{l_{\rm MAX}} (2l+1)[|1 - S_l^j|^2 + (1 - |S_l^j|^2)], \quad (6) \end{aligned}$$

where $q_e^j(E)$ and $q_a^j(E)$ are elastic and absorption cross sections of the jth atom in the molecule, respectively. S_l^j is the lth complex scattering matrix element of the jth atom in the molecule, which is related with the partial wave shift as $S_l^j = \exp(2{\rm i}\delta_{ij})$. To obtain S_l^j, we solve the following radial equation:

$$\left(\frac{{\rm d}^2}{{\rm d}r^2} + k^2 - V_{\rm opt} - \frac{l(l+1)}{r^2}\right) u_l(r) = 0. \quad (7)$$

Under the boundary condition $r \to \infty$, we obtain the asymptotic solution of Eq.(7),

$$\begin{aligned} u_l(r) \sim & kr[j_l(kr) - {\rm i}n_l(kr)] \\ & + S_l kr[j_l(kr) + {\rm i}n_l(kr)], \quad (8) \end{aligned}$$

where j_l and n_l are spherical Bessel and Neumann functions, respectively. The limit $l_{\rm MAX}$ is taken, which is enough to generate the higher partial-wave contributions until a convergence of less than 0.5% is again achieved in the TCS calculation.

Values of the parameter R in target molecules (O_2, H_2O and CH_4) are obtained by using the atomic charge density, determined from the well-known Hartree–Fock atomic wavefunctions.[8]

3. Calculations and discussion

Employing the AR model together with the complex optical model potential modified and unmodified by incorporating the concept of bonded atom, we have calculated the TCSs for positron scattering from molecules O_2, H_2O and CH_4 in the energy range from 30 to 3000eV. The modified and the unmodified TCS results, together with the available experimental and theoretical data[5–7,12–15] are given in Figs.1–3 and Table 1, respectively.

Figure 1 shows TCSs for positron scattering from O_2 together with the available experimental data obtained by Charlton *et al*[12] in the energy range from 30 to 600eV and Dababneh *et al*[13] in the energy range from 30 to 600eV, and the theoretical results obtained by Baluja and Jain[5] in the energy range from 30 to 3000eV, Raj[6] in the energy range from 100 to 500eV and Reid and Wadehra[7] in the energy range from 100 to 3000eV. From Fig.1, we can see that the modified calculations are in excellent agreement with all the experimental data[12,13] and the theoretical results obtained by Baluja and Jain[5] in the corresponding energy range. For example, the differences between the modified calculations and the experimental data obtained by Charlton *et al*[12] are about 15.6% at 50eV and 9.0% at 100eV. And the differences between the modified calculations and the data obtained by Dababneh *et al*[13] are only about 8.8% at 30.2eV and 13.3% at 60.4eV, too. Obviously, the modified calculations are much closer to the experimental data[12,13] than the theoretical results obtained by Raj[6] and Reid and Wadehra.[7]

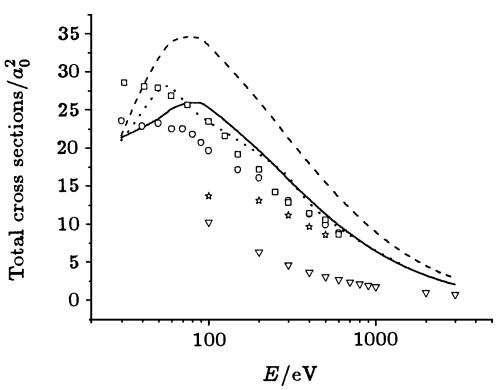


Fig.1. Total cross sections for e^+–O_2 scattering. Theoretical results: solid line, present modified results; dashed line, present unmodified results; dotted line, Baluja and Jain;[5] triangle, Reid and Wadehra;[7] star, Raj[6] Experimental results: square, Charlton *et al*;[12] circle, Dababneh *et al.*[13]

Large differences exist between the modified calculations and the experimental data obtained by Dababneh *et al*[13] at 100–300eV. And the largest one reaches 30.2%. Fortunately, the difference decreases quickly as the incident energy of electron increases. For example, it is 21.1% at 300eV, but goes down to 16.3% at 400eV.

From Fig.1, we can easily see that the results of unmodified calculations are much higher than that of the modified calculations and all the experimental data and theoretical results over a wide energy range. Thus we can say that using the AR model together with the complex optical model potential which takes into consideration the overlapping effect of electron clouds can give better TCS calculations.

Figure 2 shows TCSs for positron scattering from H_2O together with the experimental data obtained by

Sueoka and Mori[14] in the energy range from 30 to 400eV and the theoretical results obtained by Baluja and Jain[5] in the energy range from 30 to 3000eV. Very obviously, there is an excellent agreement between the modified calculations and the experimental data obtained by Sueoka and Mori[14] and the theoretical results obtained by Baluja and Jain[5] in the corresponding energy range. But there still exist large differences between the unmodified calculations and the above-mentioned results.

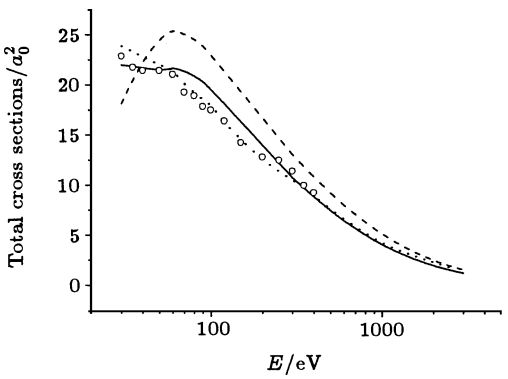


Fig.2. Total cross sections for e^+-H_2O scattering. Theoretical results: solid line, present modified results; dashed line, present unmodified results; dotted line, Baluja and Jain.[5] Experimental results: circle, Sueoka and Mori.[14]

As to the TCSs of positron–CH_4 scattering, experimental data obtained by Charlton *et al*[12] in the energy range from 30 to 600eV, Dababneh *et al*[13] in the energy range from 30 to 500eV and Sueoka and Mori[15] in the energy range from 30 to 400eV, and the theoretical results obtained by Baluja and Jain[5] in the energy range from 30 to 3000eV are shown in Fig.3. In experiments, the modified calculations are in good agreement with the data obtained by Charlton *et al*[12] in the corresponding energy range from 30 to 600eV, but are in slightly poorer agreement with the data obtained by Dababneh *et al*[13] below 300eV. In theories, the modified calculations are in excellent agreement with the results obtained by Baluja and Jain[5] in the corresponding energy range from 30 to 3000eV. In other words, the modified calculations are encouraging, but the unmodified ones are not.

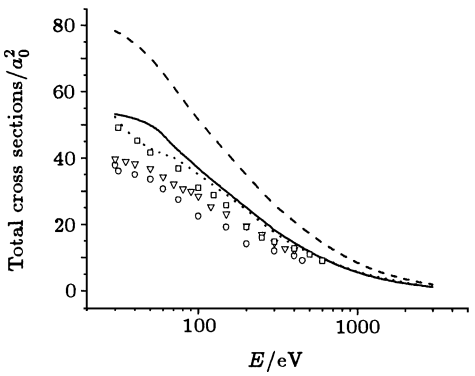


Fig.3. Total cross sections for e^+-CH_4 scattering. Theoretical results: solid line, present modified results; dashed line, present unmodified results; dotted line, Baluja and Jain.[5] Experimental results: square, Charlton *et al*;[12] circle, Dababneh *et al*;[13] triangle, Sueoka and Mori.[15]

Table 1. Total cross sections for positron scattering from O_2, H_2O and CH_4 with modified potential in units of a_0^2.

Energy/eV	O_2	H_2O	CH_4	Energy/eV	O_2	H_2O	CH_4	Energy/eV	O_2	H_2O	CH_4
30.0	21.37	21.99	53.26	500.0	11.32	7.50	11.90	1600.0	4.12	2.57	3.35
40.0	22.72	21.84	52.10	600.0	9.88	6.49	10.01	1700.0	3.87	2.41	3.11
50.0	23.55	21.28	50.86	700.0	8.76	5.70	8.57	1800.0	3.65	2.27	2.90
60.0	25.12	21.70	47.14	800.0	7.85	5.07	7.45	1900.0	3.45	2.14	2.71
70.0	25.85	21.49	43.50	900.0	7.09	4.56	6.56	2000.0	3.27	2.02	2.54
80.0	26.06	20.99	41.15	1000.0	6.46	4.13	5.83	2200.0	2.95	1.81	2.25
90.0	25.93	20.35	39.14	1100.0	5.93	3.77	5.24	2400.0	2.68	1.64	2.02
100.0	25.60	19.67	36.49	1200.0	5.46	3.46	4.73	2600.0	2.45	1.50	1.92
200.0	19.99	14.01	25.33	1300.0	5.06	3.19	4.31	2800.0	2.25	1.37	1.62
300.0	15.84	10.79	18.52	1400.0	4.71	2.96	3.94	3000.0	2.08	1.26	1.46
400.0	13.21	8.85	14.54	1500.0	4.40	2.76	3.63				

At low energies, as seen from Figs.1–3, the modified calculations substantially exceed the experimental data because we have ignored the multi-centre scattering and the valence-bond effect[16] in the AR model. But with the increasing of the incident energy, the wavelength of the incident positron becomes smaller and smaller compared with the bond length of the molecule. Then the valence-bond effect and shielding

effect contribute less and less. Therefore, the higher the energy is, the better the modified calculations are. At above 200–300eV, the contribution of molecular structure and shielding effect to the TCSs may be neglected. So, the AR model and the complex optical model potential modified by the concept of bonded atom can give excellent calculation results within the experimental errors, as verified in Figs.1–3.

4. Conclusions

The AR model neglects molecular geometry and its nuclear motion, so the molecular scattering is reduced to the atomic scattering problem which is easier to treat. The modified calculations are in good agreement with all the available measurements, even though we ignore the molecular multi-centre nature (in the case of polyatomic and hetero-nuclear molecules). It is shown that the AR model together with the complex optical model potential modified by taking into consideration the overlapping effect of electron clouds between two atoms in a molecule can be successfully used to calculate the TCSs of positron–molecule scattering above 30eV. Since the contributions from the interference occurring between the scattering amplitudes originating from the different constituent atoms of the molecule and molecular shielding effect are not included in the AR model, the results of TCS using the rule show large differences as high as 30.2%.

References

[1] Sueoka O, Makochekanwa C and Kawate H 2002 *Nucl. Instrum. Methods Phys. Res.* B **192** 206

[2] Kawada M K, Sueoka O and Kimura M 2000 *Chem. Phys. Lett.* **330** 34

[3] Liu Y F, Zhu Z L, Cong S L and Han K L 2003 *Chin. Phys.* **12** 750

[4] Ratnavelu K and Rajagopa K K 1999 *J. Phys.* B **32** L381

[5] Baluja K L and Jain A 1992 *Phys. Rev.* A **45** 7838

[6] Raj D 1993 *Phys. Lett.* A **174** 304

[7] Reid D D and Wadehra J M 1999 *Chem. Phys. Lett.* **311** 385

[8] Clementi E and Roetti C 1974 *At. Data Nucl. Data Tables* **14** 177

[9] Shi D H, Sun J F, Zhu Z L and Liu Y F 2005 *Chin. Phys.* **14** 120

[10] Staszewske G, Schwenka D W, Hirumalai D and Truhlar D G 1983 *Phys. Rev.* A **28** 2740

[11] Shi D H, Sun J F, Yang X D, Zhu Z L and Liu Y F 2004 *Chin. Phys.* **13** 1018

[12] Charlton M, Griffith T C, Heyland G R and Wright G L 1980 *J. Phys.* B **13** L353

[13] Dababneh M S, Hsieh Y F, Kauppila W E, Kwan C K, Smith S J, Stein T S and Uddin M N 1988 *Phys. Rev.* A **38** 1207

[14] Sueoka O and Mori S 1986 *J. Phys.* B **19** L373

[15] Sueoka O and Mori S 1986 *J. Phys.* B **19** 4035

[16] Hayashi S and Kuchitsu K 1976 *Chem. Phys. Lett.* **41** 575