Total cross-sections for positron scattering by a series of molecules

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Abstract. The additivity rule is employed to obtain the total (elastic+inelastic) cross-sections for positron scattering from molecules including a number of diatomic, polyatomic molecules (H_2, N_2, HCl, CO_2, NH_3, SF_6, CH_4, C_2H_4 and C_3H_8) over an incident energy range of $10-1000$ eV. The total cross-sections (TCS) of the constituent atoms of molecules are obtained by employing a complex optical model potential (composed of static, polarization and absorption potential). The present results are compared with experimental data and other theoretical calculations, good agreement is obtained in intermediate- and high-energy region.

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1 Introduction

The total cross-sections (TCS) for positron scattering from various molecules have recently been measured for energy region from a few eV to several hundred eV in many laboratories. On the theoretical side, it is well-known that positron-molecule scattering present a more complex problem than corresponding positron-atom scattering due to the multicentre nature, the lack of a center of symmetry (in the case of polyatomic and hetero-nuclear molecules) and its nuclear motion. At intermediate and high energies ($E \geq 10$ eV), almost all inelastic channels (Positronium formation, dissociation, rotational, vibrational, excitation and ionization *etc.*) are open, which makes an *ab initio* calculation almost impossible. It is thus quite obvious that most of the exact calculations carried out on the positron molecule systems have been restricted to low energies (below 10 eV) only, which has been reviewed by Armour [1]. Therefore, simpler approximation approach for positron-molecules scattering system at intermediate- and high-energy region is very necessary. The local-spherical-complex-optical potential (SCOP) method is a simple approximation, Baluja and Jain [2] have obtained the TCS for positron scattering from several diatomic and polyatomic molecules by the use of SCOP. The SCOP was generated from the molecule wave function at the Hartree-Fock level. Though the SCOP is adequate in predicting TCS in energies above 100 eV, the task is still difficult due to the lack of available of wave functions of complex molecules.

A very simple approach, namely the additivity rule [3] has been employed to obtain the TCS for electron impact on various molecules in intermediate- and high-energy region [4–7]. The results show that the additivity rule is proper for TCS calculation of electron-molecule scattering in intermediate- and high-energy region. Raj [8] made the first application of additivity rule to obtain the total cross-sections for positron scattering by a number of molecules (O_2, CO, CO_2, SO_2, CS_2, OCS and SF_6) at $100-500$ eV.

Recently [9,10], we obtained the parameter free positron polarization and absorption potential which is successful to calculate the TCS for positron-atoms scattering system in intermediate- and high-energy region. In this paper, we employ the additivity rule and complex optical potential to obtain the TCS for positron scattering from molecules including a number of diatomic, polyatomic molecules (H_2, N_2, HCl, CO_2, NH_3, SF_6, CH_4, C_2H_4 and C_3H_8) at $10-1000$ eV.

In the next section, we provide theoretical details. The results are provided and discussed in Section 3. The final concluding remarks are made in Section 4. We use atomic units in this paper until otherwith specified.

2 Theory

The basic philosophy of the additivity rule is based on the assumption that anisotropic positron-molecule interactions don't play a significant role in shaping up the TCS of the intermediate- and high-energy positron-molecule collisions. According to the additivity rule and optical

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theorem [3], the TCS (elastic + inelastic) $Q_T(E)$ of the molecules is given by

$$Q_T(E) = \frac{4\pi}{k} Im F_m(\theta = 0) \approx \frac{4\pi}{k} Im \sum_{j=1}^{N} f_j(\theta = 0)$$

$$= \sum_{j=1}^{N} q_T^j(E) \quad \text{(the additivity rule)} \quad (1)$$

where $F_m(\theta = 0)$ is the positron-molecule scattering amplitude for forward direction; $q_T^j(E)$ and f_j are the TCS due to the jth atom of the molecule and the complex scattering amplitude for constituent atoms of the molecule, respectively, N is the total number of constituent atoms of the molecule. Here it is obvious that no molecular geometry is involved in the additivity rule. So, the molecular scattering problem is reduced to the atomic scattering problem which is easier to handle.

In the present investigation we employ the complex optical model potential method to obtain the TCS for positron scattering from constituent atoms of molecule. In this method, the interaction of the positron-atom system can be represented by a local complex optical model potential, namely

$$V_{opt}(r) = V_s(r) + V_p(r) + iV_a(r). \quad (2)$$

Thus $V_{opt}(r)$ incorporates all the important physical effects. The repulsive static potential $V_s(r)$ is calculated by using the atomic charge density determined from the well-known Hartree-Fock atomic wave functions [12]; the parameter-free positron correlation-polarization potential $V_p(r)$ is of the following form

$$V_p(r) = -\frac{\alpha}{2(r^2 + r_+^2)^2} \quad (3)$$

where the constant r_+ can be determined by letting $V_p(0) = -\alpha/2r_+^4 = V_{corr}(0)$, then $r_+ = (-\alpha/2V_{corr}(0))^{1/4}$, $V_{corr}(r)$ is the near-target region ($r \leq r_c$, r_c is the crossing radius of $V_{corr}(r)$ and $-\alpha/2r_+^4$) positron correlation polarization potential of Jain [13] (*i.e.*, Eqs. (4a–c) in Ref. [13]), and α is atomic polarizability. Thus, the $V_p(r)$ in equation (3) will have a asymptotic form $-\alpha/2r_+^4$ at large r and approach $V_{corr}(r)$ in the near-target region. Meanwhile, the $V_p(r)$ (Eq. (3)) will have a smooth form at all r. This polarization potential has been discussed and employed to calculate TCS and differential cross-sections (DCS) for positron-Ar scattering at low energy, and proved fairly successful to TCS and DCS for positron-atom scattering [9].

The imaginary part of the optical potential $V_a(r)$ is the absorption potential, which represents approximately the combined effect of all the inelastic channel (Ps formation, excitation and ionization *etc.*). Recently, by studying and analyzing the quasifree model for electron scattering which was introduced preciously by Truhlar *et al.* [14], we have obtained a positron absorption potential [10]. To be convenient for readers, here we will introduce in detail our positron absorption potential [10]. The main idea of the potential is as following.

For a local imaginary potential such as iV_a the absorption probability per unit time is $-2V_a$. The result is compared with the corresponding result from classical kinetic theory for a projectile with energy $E = P^2/2$ in a free electron gas of density $\rho(r)$. For this latter case, the absorption probability per unit time is $\rho(r)\overline{\sigma_b}(k_F, p)v$, where v is the local speed of the projectile, $\overline{\sigma_b}(k_F, p)$ is the average binary collision (between the projectile and the target electrons) cross-section for absorption producing events. Thus, we can write

$$V_a = -\frac{1}{2}\rho(r)\overline{\sigma_b}(k_F, p)v. \quad (4)$$

A general expression for $\overline{\sigma_b}(k_F, p)$ is given by Goldberger [15] as

$$\bar{\sigma}_b(k_F, p) = \frac{1}{p} \int d\mathbf{k} N(\mathbf{k}, k_F)|\mathbf{p} - \mathbf{k}|$$

$$\times \int d\mathbf{g} \frac{d\sigma_b(p_0, \hat{p}_0 \hat{p}_f)}{d\Omega} \frac{1}{p_0^2} \delta(p_0 - p_f) \Theta(p', k', k_F). \quad (5)$$

Where $N(\mathbf{k}, k_F)$ is the density per target electron in momentum space, k_F is the Fermi momenta given by $k_F(\rho(r)) = [3\pi^2\rho(r)]^{1/3}$, $\mathbf{p}$ and $\mathbf{k}$ are the laboratory-frame momenta of the incident and the target electrons before the collision, $\mathbf{p}'$ and $\mathbf{k}'$ are the final values, $\mathbf{p}_0$ and $\mathbf{p}_f$ are the initial and final momenta of the scattering electron in binary-collision frame, $\mathbf{g}$ is the momentum transfer $\mathbf{g} = \mathbf{p}' - \mathbf{p} = \mathbf{p}_f - \mathbf{p}_0$, $d\sigma_b/d\Omega$ is the differential scattering cross-section for binary collisions, and Θ is the Pauli-blocked factor that is unity for Pauli-allowed final states and zero for Pauli-blocked final states in the binary collision. If the distribution of the target electron is modeled as a quasi degenerate Fermi gas, then

$$N(\mathbf{k}, k_F) = \begin{cases} N(k_F) & \text{if } k \leq k_F \\ 0 & \text{if } k > k_F. \end{cases} \quad (6)$$

Where, $N(k_F) = \frac{3}{4\pi k_F^2}$.

In the original derivation of $V_a^-(r)$ (electron absorption potential) [14], taking into account the Pauli exclusion principle, there are two imposed conditions, (i) the final energy of the incident unbound electron is required to exceed the Fermi energy of the target, namely $p'^2 \geq k_F^2$; (ii) the final energy of the initially bound electron is required to exceed the Fermi energy by the energy gap Δ, between the ground state and the lowest excited state, namely $k'^2 \geq k_F^2 + 2\Delta$. Therefore, in the case of electron, the Pauli-blocking factor Θ can be expressed by a product of two Heaviside unit-step functions as

$$\Theta = H(k'^2 - k_F^2 - w)H(p' - k_F) \quad (7)$$

where $w = 2\Delta$, Heaviside unit-step function $H(x) = 1$ if $x > 0$, $H(x) = 0$ if $x \leq 0$.

In the case of positron impact collisions, due to the absence of the Pauli exclusion constraint, condition (i) will not be applied, but condition (ii) is still valid. Therefore, we have to remove the condition (i) from the Pauli-blocking factor Θ (Eq. (7)), allowing the positron to emerge from the binary collision with any momentum. At the same time, since the lowest inelastic scattering for positron-atom begins from positronium formation, the energy gap, Δ, is taken to be the positronium formation threshold. Thus, in the case of positron, the Θ can be expressed as:

$$\Theta = H(k'^2 - k_F^2 - w) \qquad (8)$$

where Δ is the positronium formation threshold.

Since positron and electron are distinguishable particles, the differential scattering cross-section for binary collisions (positron-electron) is given approximately by the Rutherford formula [16] as:

$$\frac{d\sigma_b(p_0, \hat{p}_0 \hat{p}_f)}{d\Omega} = \frac{4}{g^4}. \qquad (9)$$

Substituting (8, 9) into (5), We have obtained average binary collision cross-section $\overline{\sigma_b}$ expressed as

$$
\begin{aligned}
\overline{\sigma_b}(k_F, p) = {} & \frac{64\pi^2}{15p^2} N(k_F) H(p^2 - w) \\
& \times \left[\frac{5k_F^3}{w} - \frac{5k_F^3}{p^2} - \frac{2k_F^5}{p^4} + H(k_F^2 + w - p^2) \right. \\
& \left. \times \left(\frac{2(k_F^2 + w - p^2)^{\frac{5}{2}}}{p^4} \right) \right].
\end{aligned}
\qquad (10)
$$

The positron absorption potential is given by equation (4).

The $q_T^j(E)$ is obtained by the method of partial waves [17]:

$$
\begin{aligned}
q_T^j(E) &= q_e^j(E) + q_a^j(E) \\
&= \frac{\pi}{k^2} \sum_{l=0}^{l_{max}} (2l+1)[|\, 1 - S_l^j\, |^2 + (1 - |\, S_l^j\, |^2)] \quad (11)
\end{aligned}
$$

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

$$\left(\frac{d^2}{dr^2} + k^2 - V_{opt} - \frac{l(l+1)}{r^2} \right) u_l(r) = 0 \qquad (12)$$

under the boundary condition

$$u_l(kr) \sim kr[j_l(kr) - in_l(kr)] + S_l kr[j_l(kr) + in_l(kr)] \qquad (13)$$

where j_l and n_l are spherical Bessel and Neumann function separately. The limit l_{max} of equation (12) is taken to

be 50. An effective-range formula

$$\tan \delta_l = \frac{\pi\alpha k^2}{(2l+1)(2l+3)(2l-1)} \qquad (14)$$

are used to generate the higher partial-wave contributions until again a convergence of less than 0.5% is achieved in the TCS.

3 Results and discussion

In intermediate- and high-energy region, using the present optical potential, we have obtained fairly good TCS results for positron scattering by He, Ne, Ar [10], Li, Na, K and Rb [11] compared with experimental data and other theoretical results. In this paper, we firstly calculate the TCS results for positron scattering from H, C, N, O, F, S and Cl atoms by the use of present optical potential in the present energy region. Then, *via* the additivity rule (1) we obtain the TCS results of a series of molecules (H_2, N_2, HCl, CO_2, NH_3, SF_6, CH_4, C_2H_4 and C_3H_8) at $10-1000$ eV. The results, along with the available experimental data and the SCOP results of Baluja and Jain, are shown in Figures 1–9.

H_2 and N_2 are two simple homonuclear diatomic molecules. Our results are shown in Figures 1 and 2 along with various experimental data [18–20] and the SCOP result [2]. For H_2 molecule, our results are in good agreement with the measurements of Charlton *et al.* [18], Hoffman *et al.* [19] and Zhou *et al.* [20] beyond 40, 100 and 100 eV, respectively, where the differences of our results compared with these experimental results are within 14%. Beyond 70 eV, our results are also in good accord with the SCOP results [2]. The case of N_2 molecule is similar to that of H_2 molecule beyond 100 eV, the differences of our results compared with the experimental results [18,19] are within 14.6%. We firstly report the theoretical TCS for positron scattering from the polar diatomic molecule HCl in the present energy region, the results are shown in Figure 3. Beyond 70 eV, it is in fairly good agreement between our results and the experimental results of Hamada *et al.* [21], the maximum discrepancy is only 7.2%.

In Figure 4, we display the TCS values for CO_2, along with the experimental results of Charlton *et al.* [18] and Kwan *et al.* [22] and the SCOP results [2]. CO_2 molecule is a simple linear polyatomic. Our results and the SCOP results are all in excellent agreement with the two measurements above 100 eV. The maximum discrepancy between our results and experimental data is 12.7%.

For NH_3, the polar polyatomic molecule, our results, SCOP results of Baluja and Jain [2] and experimental results of Sueoka *et al.* [23] are shown in Figure 5. The situation is not as good as the former several molecules (H_2, N_2, HCl and CO_2), but beyond 200 eV, our results are still excellent agreement with the experimental data and SCOP results, the maximum discrepancy between our results and experimental data [23] is 10.8%.
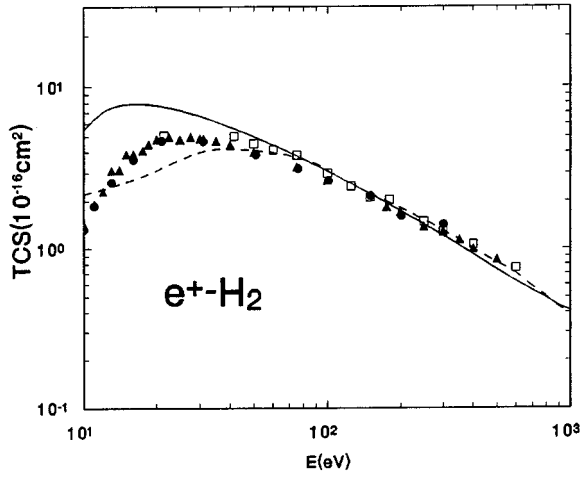
Fig. 1. Total cross-sections (10^{-16} cm^2) for positron (e$^+$)–H$_2$ scattering. Solid curve,the present results; dash curve, the SCOP results [2]. The experimental results, ($\bullet$), Zhou *et al.* [20]; ($\blacktriangle$), Hoffman *et al.* [19]; ($\square$), Charlton *et al.* [18].
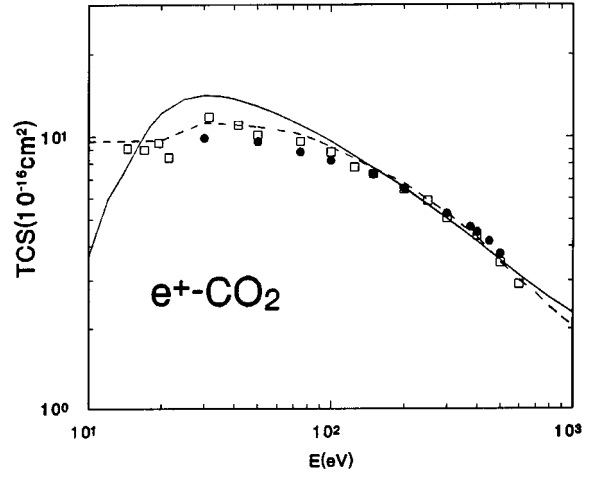

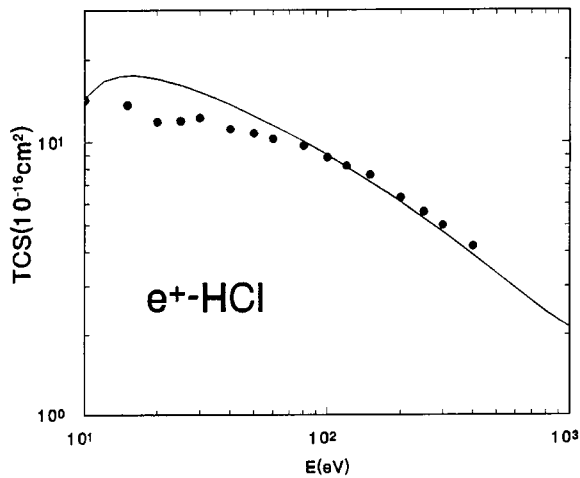**Fig. 2.** Same as in Figure 1, but for N$_2$ case, ($\bullet$), Hoffman *et al.* [19].

Fig. 3. Total cross-sections (10^{-16} cm^2) for positron (e$^+$)–HCl scattering. Solid curve,the present results. The experimental results, ($\bullet$), Hamada *et al.* [21].
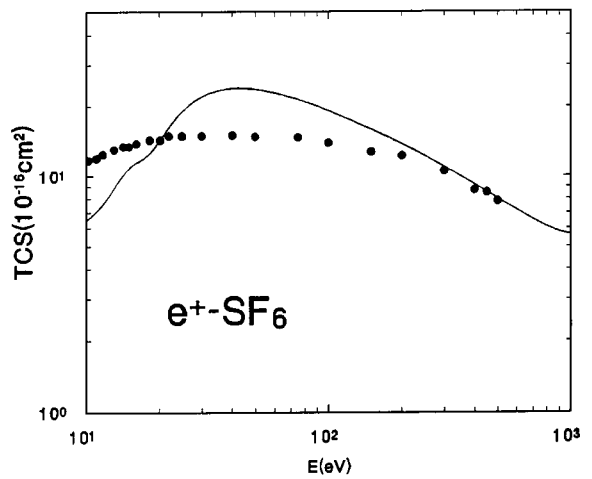

Fig. 4. Same as in Figure 1, but for CO$_2$ case, ($\bullet$), from the experimental results of Kwan *et al.* [22].

Fig. 5. Same as in Figure 1, but for NH$_3$ case. The experimental results, ($\bullet$), Sueoka *et al.* [23].

Fig. 6. Same as in Figure 3 but for SF$_6$ case. The experimental results, ($\bullet$), Dababneh *et al.* [24].
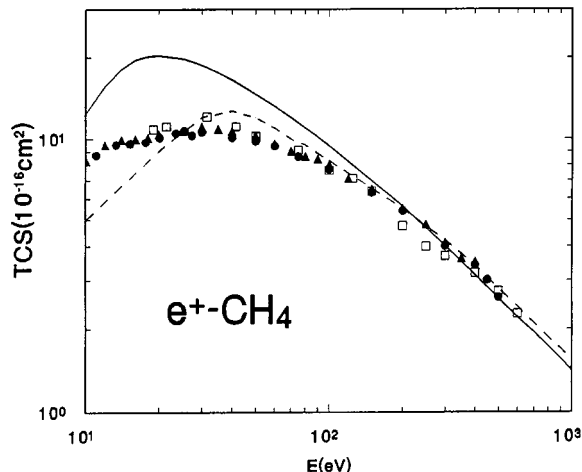
Fig. 7. Same as in Figure 1 but for CH_4 case, the experimental results, (●), Dababneh *et al.* [24], (▲), Sueoka *et al.* [26].
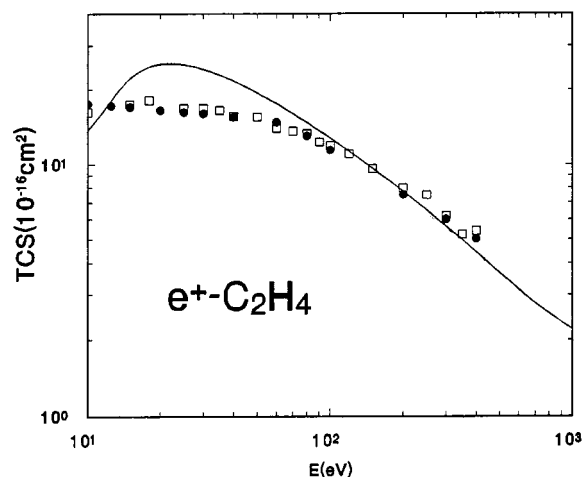

Fig. 8. Total cross-sections (10^{-16} cm^2) for positron (e^+)–C_2H_4 scattering. Solid curve, the present results. The experimental results, (●), Floeder *et al.* [25]; (□), Sueoka *et al.* [26].
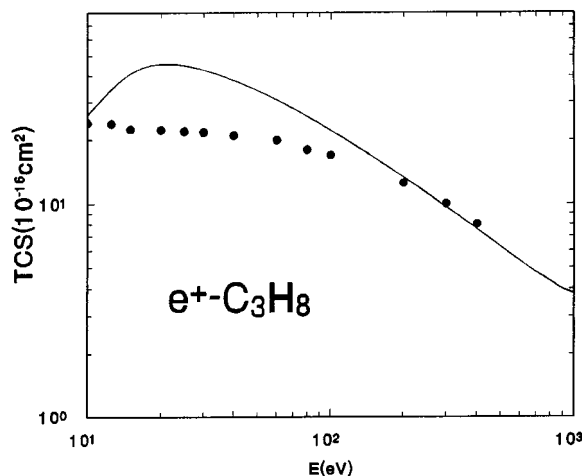

Fig. 9. Same as in Figure 8 but for C_3H_8 case.

Very little theoretical work has been done on the positron-SF_6 scattering system, because the SF_6 molecule is a very large assembly of atoms containing bound electrons, which makes difficult to handle by *ab initio* computational methods. In addition, because of the rather large nuclear charges of the fluorine atoms away from the center-of-mass, the SCOP method of Jain *et al.* by the single-center approach [2] is also unlikely to work. From this standpoint, the additivity rule will prove to be very significant for theoretical and experimental work. Our results by using the additivity rule and present optical potential are shown in Figure 6 along with the experimental results of Dababneh *et al.* [24]. Above 200 eV, the differences of our results compared with the experimental results are within 13%.

In Figures 7–9, we display the TCS values for three hydrocarbons. For CH_4 molecule we also display the SCOP results [2] along with the experimental results of Charlton *et al.* [18], Dababneh *et al.* [24] and Sueoka *et al.* [26]. The CH_4 belongs to T_d symmetry molecule. We notice that our results are in good agreement with these experimental results above 150 eV, where the maximum discrepancy is 11.7%. At low energies, our results overestimate the experimental points and SCOP results. For C_2H_4, our results agree within 12% with the experimental results of Floeder *et al.* [25] and Sueoka *et al.* [26] beyond 100 eV. For the more complex hydrocarbon C_3H_8, the situation becomes worse, but above 200 eV our results are still in excellent agreement with the experimental results of Floeder *et al.* [25]. The SCOP results and other theoretical results for C_2H_4 and C_3H_8 are not found on literatures.

From Figures 1–9, we notice that the present situations similar to the electron-molecule case [5–7]. At low energies, the present results are substantially not agreement with the experimental results because we have ignored the multicentre scattering and the valence-bond effect in the additivity rule. In addition, a close-packed molecule is not fully transparent for low energy positrons and "inner" atoms are shielded by the "outer" atoms and do not contribute to the molecular TCS [27], for diatomic and simple linear polyatomic molecules, the atomic binding and shielding effect contribute less. Therefore, the present TCS results of diatomic and simple linear polyatomic such as H_2, N_2, HCl and CO_2 are better than those of the nonlinear polyatomic molecules NH_3, SF_6, CH_4, C_2H_4 and C_3H_8.

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, in intermediate- and high-energy range (above $100-200$ eV), the contribution of molecular structure and shielding effect on the TCS may be neglected, the additivity rule and optical potential can obtain so good TCS results that the errors caused by them are within the experimental errors, as verified in Figures 1–9.

4 Conclusion

In this paper, we have extended the additivity rule and optical potential method to positron-molecule scattering system and obtained quite encouraging TCS results for a series of molecules (H_2, N_2, HCl, CO_2, NH_3, SF_6, CH_4, C_2H_4 and C_3H_8) in the intermediate- and high-energy range (beyond 100–200 eV). The results further show that the additivity rule and optical potential model are a successful approach to obtain the TCS for positron-molecule system. We can obtain conveniently the TCS results for positron scattering from all kinds of atoms by the use of optical potential model, then *via* the additivity rule, obtain the TCS results for more complex molecules in intermediate- and high-energy range (above 200 eV). This will be fairly significant for the comparisons of existing experimental work and future laboratory investigations with theoretical predictions.

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