Positron total cross sections for collisions with N$_2$ and CO$_2$ at 30—3000 eV

Deheng Shi (施德恒)[1,2], Yufang Liu (刘玉芳)[2],
Jinfeng Sun (孙金锋)[2,3], Zunlue Zhi (朱遵略)[2,3], and Xiangdong Yang (杨向东)[3]

[1]*College of Physics & Electronic Engineering, Xinyang Normal University, Xinyang 464000*
[2]*College of Physics & Information Engineering, Henan Normal University, Xinxiang 453007*
[3]*Institute of Atomic and Molecular Physics, Sichuan University, Chengdu 610065*

The total (elastic plus inelastic) cross sections for positron scattering from N$_2$ and CO$_2$ over the incident energy range from 30 to 3000 eV 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 account the overlapping effect of electron clouds between two atoms in a molecule, is employed to calculate the total cross section of positron-molecule scattering. The calculated total cross sections are in good agreement with those reported by experiments and other theories over a wide energy range.

OCIS codes: 020.2070, 020.0020.

It is well known that positron-molecule collision process plays an important role in several fields of physics. Thus a large variety of experimental investigations of positron-molecule scattering have been made[1−5]. On the theoretical side, since almost all inelastic channels (rotation, vibration, electronic excitation, and ionization etc.) are open over the intermediate- and high-energy range, though many theoretical calculation methods about positron-molecule scattering have been proposed in the past decades, few methods can give accurate results[6,7]. Therefore most of the previous calculations on the total cross sections (TCSs) for positron-molecule scattering have been restricted to lower energies.

In order to attain the accurate TCSs for position-molecule scattering at higher energies, 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 put forward. The model firstly employed by Raj[8] obtained the TCSs for positron scattering from several molecules at 100—500 eV. Though the model can easily yield the TCSs for positron scattering from small molecules, large differences are noted between the calculated results and the experimental data at lower energies.

In this letter, employing the AR model together with the modified potentials, we have calculated the TCSs for positron scattering from N$_2$ and CO$_2$ at 30—3000 eV and obtained satisfactory results.

In the AR model, total cross section $Q_{\mathrm{T}}(E)$ of the positron scattering from the molecule containing N atoms is given by

$$Q_{\mathrm{T}}(E) = \frac{4\pi}{k} \mathrm{Im} F_{\mathrm{m}}(\theta = 0)$$
$$\approx \frac{4\pi}{k} \mathrm{Im} \sum_{j=1}^{N} f_j(\theta = 0) = \sum_{j=1}^{N} q_{\mathrm{T}}^j(E), \qquad (1)$$

where $F_{\mathrm{m}}(\theta = 0)$ is the positron-molecule scattering amplitude for forward direction; $q_{\mathrm{T}}^j(E)$ and $f_j(\theta = 0)$ are the TCS and the complex scattering amplitude for the jth constituent atom, respectively.

In the present method, the potential of a constituent atom in molecules can be expressed as

$$V_{\mathrm{opt}}(r) = V_{\mathrm{rel}}^+ + iV_{\mathrm{abs}}^+(r), \qquad (2)$$

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

Thus V_{opt} incorporates all the important physical effects. In detail, the repulsive static potential $V_{\mathrm{s}}^+(r)$ for a positron-atom scattering system is calculated by using the atomic charge density, determined from the Hartree-Fock atomic wave function[9]. The parameter-free polarization potential $V_{\mathrm{p}}^+(r)$ was given by our group[10]. It is necessary to be pointed out that the polarization potential for positron scattering is same as the one for the electron while the repulsive static potential has opposite sign. And the absorption potential $V_{\mathrm{abs}}^+(r)$ was given by Staszewske et $al.$[11].

In Eq. (1), TCS of positron-molecule scattering simply equals sum of TCSs of positron scattering from each constituent atom. Obviously, no molecular geometry is taken into account. Thus there are large differences between theories and measurements over a wide energy range (dashed lines in Figs. 1 and 2) if we directly employ Eq. (2) to calculate the TCSs for positron-molecule scattering. The reason is that many complicated interactions, such as the valence bond-distortion effect, the shielding effect, and the atomic overlapping effect, exist in the scattering process.

Here, we only take into account the atomic overlapping effect between atoms. Atomic overlapping is substantially the overlapping of electron clouds between atoms. Atoms in a molecule are all bonded atoms, which greatly differ from the free ones. The reason is that the electron clouds overlapped between atoms exhibit weaker impact than the corresponding ones of free atoms in the positron-molecule scattering process due to various reasons, such as shielding. That is to say, virtual charge density $\rho'(r)$ from a bonded atom should be lower than the atomic charge density $\rho(r)$ from a free atom. So we introduce the concept of bonded atom and modify $\rho'(r)$ of bonded one as
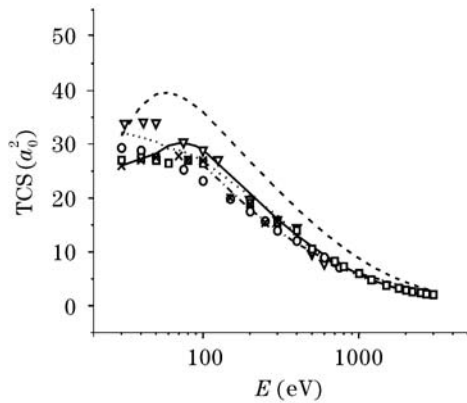
Fig. 1. TCSs for positron-N$_2$ scattering. Theoretical results: present modified calculations (solid line), present unmodified calculations (dashed line), Baluja et al.[6] (dotted line), Reid et al.[21] (dash-dotted line). Experimental results: Charlton et al.[17] (triangle), Hoffman et al.[18] (circle), Dutton et al.[19] (square), Sueoka et al.[20] (cross).
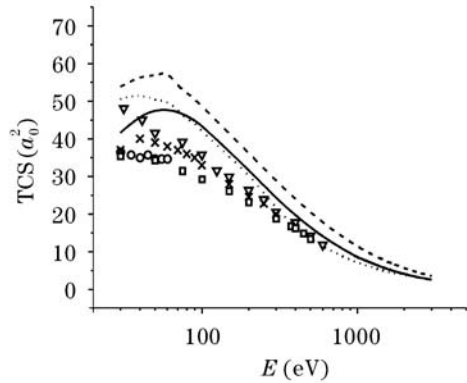

Fig. 2. TCSs for positron-CO$_2$ scattering. Theoretical results: present modified calculations (solid line), present unmodified calculations (dashed line), Baluja et al.[6] (dotted line). Experimental results: Charlton et al.[17] (triangle), Hoffman et al.[18] (circle), Sueoka et al.[20] (cross), Kwan et al.[22] (square).

$$\rho'(r) = f \cdot \rho(r), \qquad (3)$$

where f is a modification factor, which reflects the overlapping degree of electron clouds between bonded atoms. In order to attain f, three points should be considered.

1) The potential described by Eq. (2) overestimates the TCS of positron-molecule scattering at 30—3000 eV using the AR model (dashed lines in Figs. 1 and 2). Thus, in order to get accurate TCSs of positron-molecule scattering, the effect of the potential described by Eq. (2) should decrease.

2) Numerous calculations by Eq. (2) show that the more the electron number of the outmost shell in the constituent atom, the more the TCS differences between theories and experiments. When we investigate the target molecules resembling each other in structure, the results show that the more the total electron number of the constituent atom, the more the TCS differences between theories and experiments. These indicate that the modification should take into account the electron number of the outmost shell and the total electron number of

the constituent atom.

3) The atoms in molecules are quite different from those in the free states. The reason is that the atoms in molecules are bonded atoms whereas the atoms in the free state not. Thus there exists the overlapping effect of electron clouds between two atoms in a molecule, but it does not exist between the free ones. It indicates that the modification should take into account the radius of the constituent atom and the bond length between two atoms in a molecule.

According to the above-mentioned reasons, we have attained a semi-empirical expression of f

$$f = 1 - \frac{r}{R} \cdot \frac{Z}{N}, \qquad (4)$$

where r is the bond length between two atoms, R is the sum of the radii of the two atoms, Z and N are the number of electron in the outmost shell and the total number of electron in the atom, respectively. Using Eq. (4), we have attained great success in the TCS calculations of electrons scattering from about 40 molecules and the differential cross section (DCS) calculations of electrons scattering from several molecules. Some of them have been given in Refs. [12—16]. Therefore, we can conclude that the semi-empirical expression f is reasonable in the TCS and DCS calculations. Now, introducing Eq. (4) into TCS calculations for positron-molecule scattering, Eq. (2) can be modified as

$$V_{\text{opt}}(r) = V_{\text{rel}}^+ + if \cdot V_{\text{abs}}^+(r), \qquad (5)$$

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

q_{T}^j in Eq. (1) is obtained by[16]

$$\left(\frac{d^2}{dr^2} + k^2 - V_{\text{opt}} - \frac{l(l+1)}{r^2} \right) u_l(r) = 0. \qquad (6)$$

Under the boundary condition of $r \to \infty$

$$u_l(r) \sim kr[j_l(kr) - in_l(kr)] + S_l kr[j_l(kr) + in_l(kr)], \qquad (7)$$

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

Values of R in the target molecules (N$_2$ and CO$_2$) are obtained by using the atomic charge density, determined from the Hartree-Fock atomic wave function[9].

The modified and unmodified TCS results together with those obtained by experiments and other theories[6,17−22] are shown in Figs. 1 and 2, respectively.

Figure 1 shows TCSs for positron scattering from N$_2$ together with the experimental data obtained by Charlton et al.[17] at 31.5—600 eV, Hoffman et al.[18] at 30—750 eV, Dutton et al.[19] at 30—3000 eV and Sueoka et al.[20] at 30—300 eV, and the theoretical results obtained by Baluja et al.[6] at 30—3000 eV and Reid et al.[21] at 100—3000 eV. Obviously, the modified calculations are in good agreement with all the experimental data[17−20] and all the theoretical results[6,21] above 50 eV. For example, the differences between the modified calculations and the experimental data obtained by Charlton et al.[17]

are 16.6% at 50 eV and only 0.4% at 100 eV, and the ones between the modified calculations and the experimental data obtained by Sueoka et al.[20] are 11.4% at 80 eV and 7.8% at 100 eV, too. But large differences can be seen between the unmodified calculations and all the experimental data[17−20] at lower energies, especially below 500 eV.

Figure 2 shows TCSs for positron scattering from CO_2 together with the experimental data obtained by Charlton et al.[17] at 31.5—600 eV, Hoffman et al.[18] at 30—60 eV, Sueoka et al.[20] at 30—300 eV, and Kwan et al.[22] at 30—500 eV, and the theoretical results obtained by Baluja et al.[6] at 30—3000 eV. From Fig. 2, we can easily see that the modified calculations are in good agreement with the experimental data obtained by Charlton et al.[17] and Sueoka et al.[20] above 100 eV, and the theoretical results obtained by Baluja et al.[6] at 30—3000 eV (we can see that the results obtained by Baluja et al.[6] are closer to the experiments than ours at some energies, but we should note that Baluja et al.[6] employed a very complicated method in their TCS calculations.), but are in slightly poorer agreement with the experimental data obtained by Charlton et al.[17], Hoffman et al.[18], and Sueoka et al.[20] below 100 eV, and Kwan et al.[22] at lower energies. However it is necessarily noted that the ones obtained by Hoffman et al.[18] at 30—60 eV and Kwan et al.[22] at lower energies are much smaller than those obtained by Charlton et al.[17] and Sueoka et al.[20]. Very obviously, there still exist large differences between the unmodified calculations and the above-mentioned results at lower energies.

At lower energies, as shown in Figs. 1 and 2, the modified calculations are still higher than all the experimental data because we have ignored the multi-centre scattering, the shielding effect, and the valence-bond distortion effect[23] in the AR model. In addition, a close-packed molecule is not fully transparent for low energy positrons and one atom is partly shielded by others when scattering takes place. So, it is obvious that the target molecule contains more atoms and the TCS calculations show worse because both the valence-bond distortion effect and the shielding effect yield more contributions to the TCS of positron-molecule scattering. It is the reason why the TCS calculations of positron-CO_2 scattering show worse than those of positron-N_2 scattering.

As shown in Figs. 1 and 2, present modified calculations are obviously closer to the experiments than the unmodified ones over a wide energy range, especially at lower energies. The reason is that the modified potential takes into account the overlapping effect of electron clouds between two atoms in a molecule, but the unmodified potential does not. That is to say, for the AR model with the unmodified potential, the overlapped clouds in polyatomic molecules are still used to calculate the TCS of the constituent atom.

With the increase 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 distortion effect and the shielding effect contribute less and less. Therefore, the higher the energy is, the better the modified calculations are. And above 200—300 eV, 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 obtain excellent calculated results, as verified in Figs. 1 and 2.

The AR model neglects the molecular geometry and its nuclear motion, so the positron-molecule scattering is reduced to the positron-atom scattering problem. The modified calculations are encouraging, even though we ignore the molecular geometry and its nuclear motion.

References

1. O. Sueoka, C. Makochekanwa, and H. Kawate, Nucl. Instr. and Meth. in Phys. Res. B **192**, 206 (2002).
2. O. Sueoka, H. Takaki, A. Hamada, H. Sato, and M.Kimura, Chem. Phys. Lett. **288**, 124 (1998).
3. M. K. Kawada, O. Sueoka, and M. Kimura, Chem. Phys. Lett. **330**, 34 (2000).
4. C. Makochekanwa, O. Sueoka, and M. Kimura, Phys. Rev. A **68**, 032707 (2003).
5. H. Tanaka, Y. Tachibana, M. Kitajima, O. Sueoka, H. Takaki, A. Hamada, and M. Kimura, Phys. Rev. A **59**, 2006 (1999).
6. K. L. Baluja and A. Jain, Phys. Rev. A **45**, 7838 (1992).
7. M. J. Brunger and S. J. Buckman, Phys. Rep. **357**, 215 (2002).
8. D. Raj, Phys. Lett. A **174**, 304 (1993).
9. E. Clementi and C. Roetti, Atomic Data and Nuclear Data Tables **14**, 177 (1974).
10. X. Zhang, J. Sun, and Y. Liu, J. Phys. B **25**, 1893 (1992).
11. G. Staszewske, D. W. Schwenka, D. Thirumalai, and D. G. Truhlar, Phys. Rev. A **28**, 2740 (1983).
12. D.-H. Shi, Z.-L. Zhu, J.-F. Sun, X.-D. Yang, Y.-F. Liu, and Y. Zhao, Chin. Phys. Lett. **21**, 474 (2004).
13. D.-H. Shi, J.-F. Sun, X.-D. Yang, Z.-L. Zhu, and Y.-F. Liu, Chin. Phys. **13**, 1018 (2004).
14. J. Sun, D. Shi, Z. Zhu, and Y. Liu, Chin. Opt. Lett. **1**, 624 (2003).
15. D.-H. Shi, Y.-F. Liu, J.-F. Sun, X.-D. Yang, and Z.-L. Zhu, Chin. Phys. Lett. **22**, 321 (2005).
16. D. Shi, J. Sun, X. Yang, Z. Zhu, and Y. Liu, Chin. Opt. Lett. **3**, 1 (2005).
17. M. Charlton, T. C. Griffith, G. R. Heyland, and G. L. Wright, J. Phys. B **13**, L353 (1980).
18. K. R. Hoffman, M. S. Dababneh, Y.-F. Hsieh, W. E. Kauppila, V. Pol, J. H. Smart, and T. S. Stein, Phys. Rev. A **25**, 1393 (1982).
19. J. Dutton, C. J. Evans, and H. L. Mansour, J. Phys. B **20**, 2607 (1987).
20. O. Sueoka and S. Mori, J. Phys. Soc. Jpn. **53**, 2491 (1984).
21. D. D. Reid and J. M. Wadehra, Chem. Phys. Lett. **311**, 385 (1999).
22. Ch. K. Kwan, Y.-F. Hsieh, W. E. Kauppila, S. J. Smith, T. S. Stein, M. N. Uddin, and M. S. Dababneh, Phys. Rev. A **27**, 1328 (1983).
23. D. Raj and S. Tomar, J. Phys. B **30**, 1989 (1997).