

Electron impact total (elastic + inelastic) cross sections with simple molecules consisting of N & O atoms at 100 – 1600 eV

Jinfeng Sun (孙金锋)¹, Deheng Shi (施德恒)^{1,2}, Zunlue Zhu (朱遵略)¹, and Yufang Liu (刘玉芳)¹

¹Department of Physics, Henan Normal University, Xinxiang 453002

²Department of Foundation, The First Aeronautical College of Air Force, Xinyang 464000

Received September 5, 2003

A model complex optical potential rewritten by the conception of bonded atom, which considers the overlapping effect of electron cloud, is employed to calculate the total (elastic + inelastic) cross sections with simple molecules (N₂, O₂, NO₂, NO, N₂O) consisting of N & O atoms over an incident energy range of 100 – 1600 eV by the use of additivity rule at Roothaan-Hartree-Fock level. In the study, the complex optical potential composed of static, exchange, correlation polarization plus absorption contributions firstly uses bonded-atom conception. The qualitative results are compared with experimental data and other calculations wherever available and good agreement is obtained. The total cross sections of electron-molecule scattering above 100 eV can be successfully calculated.

OCIS codes: 020.2070, 020.0020.

It is well known that electron-molecule scattering presents a more complex problem than the corresponding electron-atom scattering due to the multicentre nature, the lack of a centre of symmetry (in the case of polyatomic and hetero-nuclear molecules) and its nuclear motion. Hence, for electron-molecule scattering, many different approaches have been proposed^[1,2]. Here, in the intermediate- and high-energy region, since acquiring molecular wavefunction is very difficult, where almost all inelastic channels (rotational, vibrational, electronic excitation and ionization process, etc.) are open, a conventional close-coupling theory for electron-molecule scattering is an arduous task and almost impossible to carry out. It is, therefore, not surprising that many previous calculations on the total cross section (TCS) for electron-molecule scattering have been restricted to low-energy region.

Recently, a fairly simple approach, namely the additivity rule^[3], was employed to obtain the TCS for the electron-molecule scattering. According to the additivity rule, the TCS $Q_T(E)$ of the molecules is given by

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

where $q_T^j(E)$ and f_j are the TCS due to the j th atom of the molecule and complex scattering amplitude for constituent atoms of the molecule, respectively. Here it is obvious that no molecular geometry is involved in the additivity rule. So, the molecular scattering problem is reduced to the atomic problem which is easier to handle.

In the present investigation the atoms of molecule are replaced by the appropriate complex optical potential

$$V_{\text{opt}}(r) = V_s(r) + V_p(r) + V_e(r) + iV_\alpha(r). \quad (2)$$

Thus $V_{\text{opt}}(r)$ incorporates all the important physical effects. In detail the static potential $V_s(r)$ for e-atom

systems is calculated by using the atomic charge density, determined from the well-known Roothaan-Hartree-Fock atomic wavefunctions^[4]. Exchange potential $V_e(r)$ provides a semiclassical energy-dependent form of Riley and Truhlar^[5]. Polarization potential $V_p(r)$ was given by our group^[6], which has a correct asymptotic form $-\alpha/2r^4$ at large r and approaches the free-electron-gas correlation energy $V_{\text{CO}}(r)$ proposed by Perdew in the near-target region

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

where the constant r_{CO} can be determined by letting $V_p(0) = -\frac{\alpha}{2r_{\text{CO}}^4} = V_{\text{CO}}(r=0)$ and α is atomic polarizability. The absorption potential $V_\alpha(r)$ ^[7] represents the combined effect of all the inelastic channels. It is written as

$$V_\alpha(r) = -\rho(r)(T_L/2)^{1/2}(8\pi/5k^2k_F^3) \\ \times H(k^2 - k_F^2 - 2\Delta)(A + B + C), \quad (4)$$

where

$$T_L = k^2 - V_s - V_e - V_p, \\ A = 5k_F^3/2\Delta, \quad B = -k_F^3(5k^2 - 3k_F^2)/(k^2 - k_F^2)^2, \\ C = 2H(2k_F^3 + 2\Delta - k^2) \frac{(2k_F^2 + 2\Delta - k^2)}{(k^2 - k_F^2)^2},$$

where Δ is the mean excitation energy; $\rho(r)$ is the atomic charge density; k and k_F are the energy of incident electron and Fermi momentum, respectively; $H(x)$ is a Heaviside function defined by $H(x) = 1$ for $x \geq 0$, and $H(x) = 0$ for $x < 0$.

In the previous investigation the additivity rule did not consider the formation of the chemical bond and overlapping effect of electron cloud. So, calculation errors are great at lower energy range. In addition, calculations have been seldom done when incident energy is above

1000 eV due to solution divergence of differential equation.

Here, we introduce the conception of bonded atom and consider the overlapping effect of electron cloud. That is to say, atom in molecule is different from the one in free state. Thus, electron charge density $\rho(r)$ must be rewritten as

$$\rho'(r) = f \cdot \rho(r), \quad (5)$$

$$f = 1 - \frac{r}{R} \cdot \frac{Z}{N}, \quad (6)$$

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

$$V_{\text{opt}}(r) = f \cdot V_s(r) + V_p(r) + V_e(r) + if \cdot V_a(r). \quad (7)$$

$q_T^j(E)$ in Eq. (1) is obtained by the method of partial waves

$$\begin{aligned} q_T^j(E) &= q_e^j(E) + q_a^j(E) \\ &= \frac{\pi}{k^2} \sum_{l=0}^{l_{\text{MAX}}} (2l+1) \times [1 - |S_l^j|^2 + (1 - |S_l^j|^2)], \quad (8) \end{aligned}$$

where $q_e^j(E)$ and $q_a^j(E)$ are elastic and absorption cross sections, respectively; S_l^j is the complex scattering matrix element of the j th 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

$$\left(\frac{d^2}{dr^2} + k^2 - V_{\text{opt}} - \frac{l(l+1)}{r^2} \right) u_l(r) = 0 \quad (9)$$

under the boundary condition ($r \sim \infty$)

$$u_l(kr) \sim kr[j_l(kr) - in_l(kr)] + S_l kr[j_l(kr) + in_l(kr)], \quad (10)$$

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

In this letter, employing the additivity rule along with the complex optical potential model rewritten by the conception of bonded atom, we have calculated the TCS for e-molecules (N_2 , O_2 , NO_2 , NO , N_2O) consisting of N & O atoms scattering over an incident energy range of 100 – 1600 eV. The reason of selecting molecules N_2 , O_2 , NO_2 , NO , N_2O as targets is two-fold. Firstly, All of these molecules have experimental data^[8–14] in the wide energy range and secondly, the structure of these targets is not too complicated. Thus calculation is easily made.

Values of parameters R in molecules N_2 , O_2 , NO_2 , NO , N_2O are achieved by using the atomic charge density, determined from the well-known Roothaan-Hartree-Fock atomic wavefunction^[4]. Atomic units are employed throughout this paper.

The present results of TCS at 100 – 1600 eV along with available experimental and theoretical data are given in

Figs. 1 – 5. Now we start by discussing TCS for e- O_2 scattering. Figure 1 shows the e- O_2 TCS present results (real line) along with the available data of Dalba group^[8] (triangle) and Dababneh group^[9] (squares). From Fig. 1 we notice that the present values of TCS resulting from the use of the additivity rule along with the complex optical potential model rewritten by the conception of bonded atom is in excellent agreement and is only at most 7.7% higher than the measurements of Dalba *et al.* and Dababneh *et al.* at 100 – 1600 eV. Figure 2 shows e- NO TCS together with the experimental results of Dalba group^[8] (circle) and the theoretical results of Joshipura group^[10] (square). The present values (real line) are at most 10% slightly higher than the experimental data^[8] but only 4.0% larger than Joshipura group^[10] at lower energies. And the said overestimation rapidly decreases as energy increases. So, the overall results are excellent.

Figures 3 and 4 show e- N_2 and e- NO_2 TCS (real line) separately. From Fig. 3 we notice that the overestimation as seen is at most 23% higher than Dalba group^[8] (triangle) and Blaauw group^[11] (star) in the incident energy range of 200 – 250 eV but 16% lower than in the

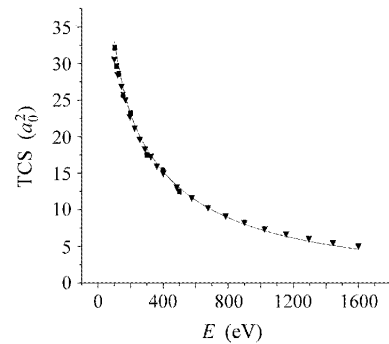


Fig. 1. Total cross section for e- O_2 scattering.

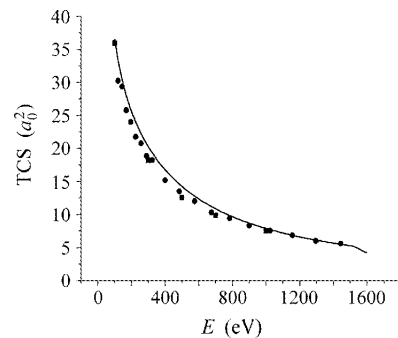


Fig. 2. Total cross section for e- NO scattering.

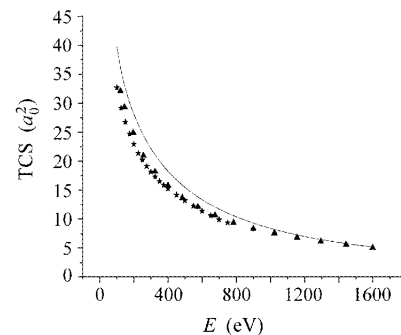
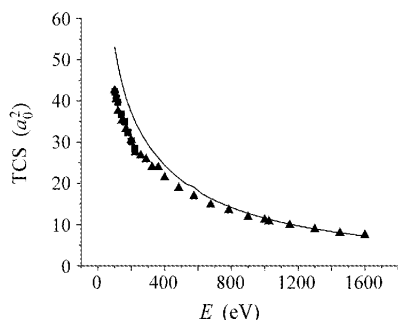


Fig. 3. Total cross section for e- N_2O scattering.

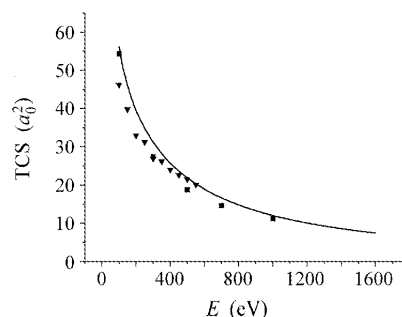
Fig. 4. Total cross section for e-NO₂ scattering.

incident energy range of 100 – 200 eV and 250 – 1600 eV and rapidly decreases as energy increases. So, we guess the larger discrepancies in the incident energy range 200 – 250 eV are mainly caused by experimental errors^[8]. Figure 4 also shows at most 29% overestimation by Zecca group^[12] (triangle) and Szmytkowski group^[13] (squares) at 121 eV. But when energy is above 300 eV, the discrepancies are at most 15%. This shows that the additivity rule along with the complex optical potential model rewritten by the conception of bonded atom appears to succeed above 300 eV for e-NO₂. The overestimation is due to the neglect of multiple scattering as well as valence-bond effects. Though we introduce the conception of bonded atom, the correlation is not enough large for two-double-bond molecules such as NO₂.

Especially, Zecca group^[12] have already pointed out that their experimental errors are 2.34 (5.5%), 1.57 (3.5%) and 2.06 (5.5%) in unit of a_0^2 at 100, 110 and 121 eV, separately. And in the incident energy range of 144 – 200 eV, the relative experimental errors are at least 2.2%. If we consider this factor within the combined uncertainties, even in the incident energy range of 100 – 220 eV, the discrepancies beyond few incident energy points may also be below 20%. The same analysis is for the experimental results of Szmytkowski group^[13].

Figure 5 shows e-N₂O TCS along with the experimental results of Kwan *et al.*^[14] (squares) and the theoretical results of Joshipura group^[10] (triangle). The present values (real line) are somewhat larger than the measured values in the incident energy range of 100 – 300 eV whereas in excellent agreement above 300 eV. The overestimation as seen here is about 15% – 20% in the incident energy range of 100 – 200 eV but about 10% – 15% in the incident energy range of 200 – 300 eV. Overall, the present results are encouraging above 100 eV and are better than the calculation values of Joshipura group at some energy region.

Obviously, the present values show a good agreement as shown in Figs. 1 – 5 except e-NO₂ in the incident energy range of 200 – 250 eV when the experimental errors^[12] are not considered. From the above calculated, basically, the additivity rule along with the complex optical potential model rewritten by the conception of bonded atom can be used successfully to calculate the TCS of electron-molecule scattering above 100 eV, whereas the rule together with the complex optical potential model not rewritten by the conception of bonded atom is only successfully used above 300 – 500 eV^[15,16]. So, we can say that the present method has yielded encouraging TCS for simple molecules such as N₂, O₂, NO₂, NO,

Fig. 5. Total cross section for e-N₂O scattering.

N₂O consisting of N & O atoms.

However, there still exists discrepancy between our results and experimental measurements. Here, we do not include multiple scattering and shielding effect. How to include the multiple scattering and shielding effect in the theoretical calculation to solve the discrepancy between our results and experimental measurements of triple-bond and two-double-bond molecules, which is larger than the one of single-bond and single-double-bond molecules, is another problem which is still in research.

This work was supported by the National Natural Science Foundation of China under Grant No. 10174019. D. Shi is the author to whom the correspondence should be addressed, his e-mail address is shideheng@yahoo.com.cn.

References

1. A. Zecca, J. C. Nogueira, G. P. Karwasz, and R. S. Brusa, *J. Phys. B: At. Mol. Opt. Phys.* **28**, 477 (1995).
2. M.-T. Lee and I. Iga, *J. Phys. B: At. Mol. Opt. Phys.* **32**, 453 (1999).
3. D. Raj, *Phys. Lett.* **160**, 571 (1991).
4. E. Clementi and C. Roetti, *Atomic Data and Nuclear Data Tables* **14**, 177 (1974).
5. M. E. Riley and D. G. Truhlar, *J. Chem. Phys.* **63**, 2182 (1975).
6. X. Zhang, J. Sun, and Y. Liu, *J. Phys. B: At. Mol. Opt. Phys.* **25**, 1893 (1992).
7. G. Staszewska, D. W. Schwenke, D. Thirumalai, and D. G. Truhlar, *Phys. Rev. A* **28**, 2740 (1983).
8. G. Dalba, P. Fornasini, R. Grisenti, G. Ranieri, and A. Zecca, *J. Phys. B: At. Mol. Opt. Phys.* **13**, 4695 (1980).
9. M. S. Dababneh, Y.-F. Hsieh, W. E. Kauppila, C. K. Kwan, S. J. Smith, T. S. Stein, and M. N. Uddin, *Phys. Rev. A* **38**, 1207 (1988).
10. K. N. Joshipura and P. M. Patel, *Z. Phys. D* **29**, 269 (1994).
11. H. J. Blaauw, R. W. Wagenaar, D. H. Barends, and F. J. de Heer, *J. Phys. B: At. Mol. Opt. Phys.* **13**, 359 (1980).
12. A. Zecca, J. C. Nogueira, G. P. Karwasz, and R. S. Brusa, *J. Phys. B: At. Mol. Opt. Phys.* **28**, 477 (1995).
13. C. Szmytkowski, K. Maciag, and A. Krzysztowicz, *Chem. Phys. Lett.* **190**, 141 (1992).
14. Ch. K. Kwan, Y.-F. Hsieh, W. E. Kauppila, S. J. Smith, T. S. Stein, M. N. Uddin, and M. S. Dababneh, *Phys. Rev. Lett.* **52**, 1417 (1984).
15. J. Sun, Y. Jiang, and L. Wan, *Phys. Lett. A* **195**, 81 (1994).
16. Y. Liu, J. Sun, Z. Li, Y. Jiang, and L. Wan, *Z. Phys. D* **42**, 45 (1997).