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A revised additivity rule for electron scattering from ethylene, propene, butene, ethane, propane and butane

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ABSTRACT

Considering the difference between the bound atom in a molecule and the free atom, the original additivity rule is revised. Using the revised additivity rule, the total cross sections for electron scattering by ethylene (C_2H_4), propene (C_3H_6), butene (C_4H_8), ethane (C_2H_6), propane (C_3H_8) and butane (C_4H_{10}) are calculated over the energy range 10–1000 eV. The results of the revised additivity rule are compared with those obtained by experiments and the revised additivity rule can give better agreement with experimental values than the original additivity rule.

1. Introduction

Ethylene (C_2H_4), propene (C_3H_6), butene (C_4H_8), ethane (C_2H_6), propane (C_3H_8) and butane (C_4H_{10}) are hydrocarbon molecules. They play an important role in plasma processing in tokamak fusion devices and many other fields [1]. Their industrial importance has been established over the past few years [2]. Electron scattering cross sections for these molecules are indispensable parameters for simulating the above processes [3]. Numerous experimental investigations for electron scattering from these molecules have been made. Szmytkowski et al. [4], Sueoka and Mori [5] and Floeder et al. [6] have measured the total cross sections for electron scattering from C_2H_4 at energies 0.6–370 eV, 1–400 eV and 5–400 eV, respectively. Ariyasinghe and Powers [7] have reported the total cross sections of C_2H_4 for the energy range 200–1400 eV and Wickramarachchi et al. [8] have measured the total cross sections for C_2H_4 in the energy range 200–4500 eV. Total electron cross sections for C_3H_6 have been measured by Szmytkowski and Kwitnewski [9], Floeder et al. [6] and Nishimura and Tawara [10] in the energy range 0.5–370 eV, 5–400 eV and 4–500 eV, respectively. Wickramarachchi et al. [8] have obtained the experimental total cross sections for C_3H_6 for the energy range 200–4500 eV. For C_4H_8, Floeder et al. [6] have measured the total electron cross sections for 5–400 eV energy electrons and Wickramarachchi et al. [8] have reported the experimental total cross sections for the energy range 200–4500 eV. For C_2H_6, three groups, Sueoka and Mori [5], Floeder et al. [6] and Nishimura and Tawara [10] have measured the total cross sections below 500 eV. Ariyasinghe et al. [11] have reported the total cross sections for electron scattering from C_2H_6 at energies 300–4000 eV by measurement of the electron-beam intensity attenuation through a gas cell. The experimental total cross sections for electron scattering from C_3H_8 have been published by Szmytkowski and Kwitnewski [12], Floeder et al. [6] and Nishimura and Tawara [10] at energies 0.5–370 eV, 5–400 eV and 4–500 eV, respectively, and by Ariyasinghe et al. [11] for 300–4000 eV. Total cross sections for C_4H_{10} have been measured by Floeder et al. [6] at 5–400 eV and by Ariyasinghe et al. [11] at 300–4000 eV.

In theory, electron-molecule scattering presents a more complex problem than corresponding electron-atom scattering due to the multi-center nature, the lack of a center of symmetry and its nuclear motion. Many approaches have been proposed and developed. Among these approaches, the additivity rule [13] is a relatively simple but effective one. The additivity rule method is based on the assumption that anisotropic electron-molecule interactions do not play an important role in shaping up the total cross sections of the intermediate- and high-energy electron-molecule collisions. Thus, the total cross section for a molecule is the sum of the total cross sections for the constituent atoms. Raj [14] made the first application of the additivity rule to obtain the elastic cross sections for electron scattering from simple molecules. Joshipura and Patel [15] and Sun et al. [16] also employed the additivity rule to obtain the total cross sections (elastic and inelastic) for electron scattering with simple diatomic and triatomic molecules and proved that the additivity rule is proper for the calculation of the

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total cross sections for electron scattering from simple and smaller molecules in intermediate- and high-energy range. For complex and larger polyatomic molecules, although the additivity rule can give good results at high enough energies, larger discrepancies between the additivity rule and the measurements can still be seen at lower energies [17–19]. This is because no molecular structure is considered and the electron-molecule scattering is reduced to electron-atom scattering in the additivity rule method. In this paper, considering the difference between the bound atom in a molecule and the corresponding free atom, we shall revise the additivity rule method. The total cross sections for electron scattering from ethylene (C_2H_4), propene (C_3H_6), butene (C_4H_8), ethane (C_2H_6), propane (C_3H_8) and butane (C_4H_{10}) in the energy range 10–1000 eV have been calculated with the revised additivity rule. The present results are compared with the available experimental and theoretical data.

2. Theoretical model

In the additivity rule model [14], molecule orbits can be described by the sum of the valence orbits of all atoms present in the molecule. As a result, the total cross section of electron-molecule scattering is written as the sum of the total cross sections of atoms. Thus the total cross section Q_T for molecule is given by

$$Q_T = \frac{4\pi}{k}\mathrm{Im}f_M(\theta=0) = \frac{4\pi}{k}\mathrm{Im}\sum_{j=1}^{N}f_j(\theta=0) = \sum_{j=1}^{N}q_T^j(E) \tag{1}$$

where q_T^j and f_j are the total cross section due to the jth atom of the molecule and the complex scattering amplitude for constituent atoms of the molecule, respectively. The q_T^j of Eq. (1) for the jth atom is obtained by the method of partial-waves:

$$q_T^j = \frac{\pi}{k^2}\sum_{l=0}^{l_{\max}}(2l+1)\left[\left|1-s_l^j\right|^2 + \left(1-\left|s_l^j\right|^2\right)\right] \tag{2}$$

where s_l^j is the lth complex scattering matrix element of the jth atom, which is related to the partial-wave phase shift as $s_l^j = \exp(2i\delta_{lj})$. The limit $l_{\max}$ is taken, which is enough to generate the higher partial-wave contributions until a convergence of less than 0.5% is achieved in the total cross section calculation. To obtain s_l^j we solve the following radial equation

$$\left(\frac{d^2}{dr^2} + k^2 - 2V_{\mathrm{opt}} - \frac{l(l+1)}{r^2}\right)u_l(r) = 0 \tag{3}$$

Under the boundary condition

$$u_l(kr) \sim kr[j_l(kr) - in_l(kr)] + s_l kr[j_l(kr) + in_l(kr)] \tag{4}$$

where j_l and n_l are spherical Bessel and Neumann functions separately. The atom is replaced by the complex optical potential

$$V_{\mathrm{opt}} = V_s(r) + V_e(r) + V_p(r) + iV_a(r) \tag{5}$$

It incorporates all the important physical effects. Presently the static potential $V_s(r)$ for electron-atom system is calculated from the well-known Hartree–Fock atomic wave functions [20]. Exchange potential $V_e(r)$ provides a semi-classical energy-dependent form of Riley and Truhlar [21]. Zhang et al. [22] gives a smooth form at all r for polarization potential $V_p(r)$, which has a correct asymptotic form at large r and approaches the free-electron-gas correlation potential [23] in the near-target region. This potential model has been proved fairly successful to total cross sections for electron-atom scattering [22]. The imaginary part of the optical potential $V_a(r)$ is the absorption potential, which represents approximately the combined effect of all the inelastic channels. The absorption potential is derived from a quasifree-scattering model by Staszawska et al. [24], and then modified by Jiang et al. [25]. The absorption potential of Jiang et al. [25] has been used to calculate the total cross sections for electron scattering from Ar, Kr and Xe atoms at 0.1–300 eV and obtained better agreements with experiments. Here, the absorption potential of Jiang et al. is adopted. The optical potential is dependent on the atomic charge density $\rho_0(r)$.

From the above equations, we can see that the original additivity rule model does not differentiate between the free atom and the bound atom in the molecule. Considering this, we present

$$\rho(r) = f \cdot \rho_0(r) \tag{6}$$

$\rho(r)$ is the charge density of the bound atom in the molecule and $\rho_0(r)$ is the charge density of the corresponding free atom. f is a revised factor for a bound atom in the molecule. To obtain factor f, two points should be considered:

(1) The total cross sections calculated from the optical potential with the additivity rule are much higher than the experimental results at lower energies. So, to get accurate total cross sections for electron scattering from molecules, the effect of the optical potential should decrease at lower energies.

(2) The bound atoms in a molecule are different from the corresponding free atoms. The reason is that there exits the overlapping effect of electron cloud between two atoms which form the chemical bond in a molecule. This shows that the revision should be related to the radii of the constituent atoms and the bond length between the two atoms. Between the two atoms which form the chemical bond in a molecule, the more the electron number of the constituent atom is, the smaller the influence from the overlapping effect of electron cloud is. This indicates that the revision should also have relation with the electron number of the constituent atom.

From the above two points, many f factors are tested and good agreements between the revised results and the experimental data are obtained by

$$f = 1 - \frac{R}{R+d}\frac{N-Z}{N} \tag{7}$$

where d is the bond length between two bound atoms in the molecule and R is the sum of the radii of the corresponding two free atoms. Z is the electron number in the atom and N is the sum of the electron number of the two atoms which form the chemical bond. Thus, the original additivity rule is revised with Eqs. (6) and (7). Obviously, the revised additivity rule is related to the molecular structure.

3. Results and discussion

The total cross sections for electron scattering from ethylene (C_2H_4), propene (C_3H_6), butene (C_4H_8), ethane (C_2H_6), propane (C_3H_8) and butane (C_4H_{10}) have been calculated in the energy range 10–1000 eV with the above revised additivity rule. The present results are listed in Tables 1 and 2 and also compared with the experimental and other theoretical results shown in Figs. 1–6.

In Fig. 1, our present results for C_2H_4 are compared with the measurements of Szmytkowski et al. [4], Ariyasinghe and Powers [7], Wickramarachchi et al. [8], Sueoka and Mori [5], Floeder et al. [6] and the theoretical values of Floeder et al. [6], Wickramarachchi et al. [8], Nishimura and Tawara [10], Vinodkumar et al. [26], Garcia and Manero [27], and Szmytkowski [28]. The present results are in better agreement with the experimental data than the original additivity rule results in the whole energy range. For example, the difference between the present results with the experimental data of Wickramarachchi et al. [8] is only 9% at