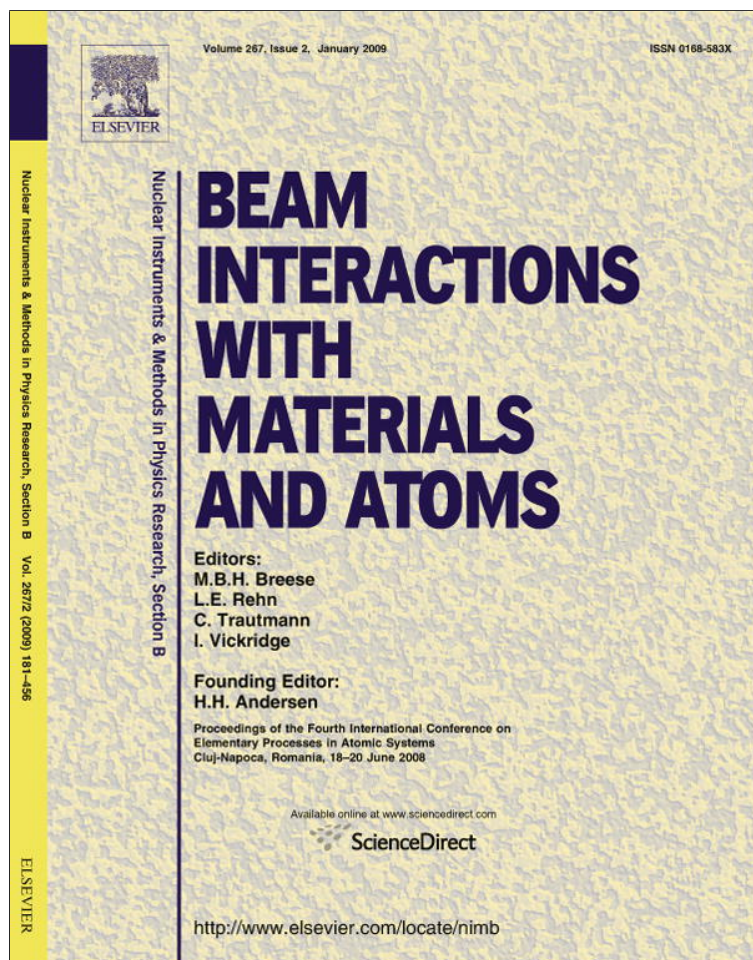


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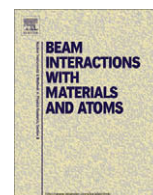
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Distorted-wave Born approximation for the ionization of molecules by positron and electron impact

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ABSTRACT

Theoretical DWBA (distorted-wave Born approximation) cross sections for the ionization of several molecules by positron and electron impact are compared. In this work we study the effect of the projectile charge sign on the ionization cross sections when positron and electron projectiles are considered. The exchange interaction effect in case of electron projectile on the cross section is also discussed.

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1. Introduction

Positron and electron impact ionization of molecules is important not only from the viewpoint of practical applications. The comparison of the measured and theoretically determined cross sections for these projectiles offers a deeper insight into the dynamics of collision processes, and guides us to a better understanding of some fundamental effects like positronium formation, annihilation or exchange interaction. Experimental data are available both for positron [1–3] and electron [4–10] impact ionization of molecules. Previously, we have calculated cross sections for the ionization of several molecules by positron and electron impact. Our earlier investigations were based on the simple CPE (Coulomb plus plane waves with full energy range) and the more complex ES (electron screening) and TS (total screening) distorted-wave models. The CPE model uses Coulomb or plane waves for the description of the incident, scattered and ejected particles. The ES and TS models describe these particles by distorted waves, calculated in the screened and spherically averaged potential created by the nuclei and electrons of the target molecule. We found that the correct description of the ejected electron is of fundamental importance in order to obtain accurate cross sections. The best agreement with experimental data was given by the ES model.

In the framework of our ES model, we compare theoretical cross sections for the ionization of a few molecules by positron and elec-

tron impact. We attempt to single out the differences between the cross sections arising from the different charge signs of the projectiles. Also, the effect of the exchange interaction on the cross section is discussed in the case of electron impact ionization.

In the present work, we have determined direct cross sections for the ionization of several molecules (N₂, CO, O₂ and CH₄) by positron and electron impact. For electron projectiles, total cross sections were also calculated by taking into account the contributions from both the direct and exchange processes.

2. Theory

The adopted theory was described elsewhere [11–17]. Here we give only a short outline of the theory, emphasizing the main steps.

The triple differential cross section for the ionization of molecules by electron or positron impact may be written as

$$\frac{d^3\sigma}{dk_f dk_e dE_e} = \sum_r \frac{(2\pi)^4}{E_i} |f_r^r|^2, \quad (1)$$

where the sum runs over all occupied molecular orbitals. $\hat{\mathbf{k}}_f$ and $\hat{\mathbf{k}}_e$ stand for the angles of the momenta of the scattered and ejected particles, respectively. The energy of the projectile was denoted by E_i , while E_e stands for the energy of the ejected electron. For positron projectile the total scattering amplitude, f_r^r , coincides with the f_d^r direct amplitude, while for electron projectile exchange effects lead to

$$|f_r^r|^2 = |f_d^r|^2 + |f_{ex}^r|^2 - \alpha |f_d^r| |f_{ex}^r|, \quad (2)$$

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where α is a phase factor [18]. The direct and exchange scattering amplitudes, f_d^r and f_{ex}^r , may be expressed as

$$f_d^r = \langle \phi_f(\mathbf{r}_1) \phi_e(\mathbf{r}_2) | V(r_{12}) | \phi_i(\mathbf{r}_1) \phi_b^r(\mathbf{r}_2) \rangle, \quad (3)$$

$$f_{ex}^r = \langle \phi_e(\mathbf{r}_1) \phi_f(\mathbf{r}_2) | V(r_{12}) | \phi_i(\mathbf{r}_1) \phi_b^r(\mathbf{r}_2) \rangle, \quad (4)$$

where ϕ_i , ϕ_f and ϕ_e are the wavefunctions of the incident, scattered and ejected particles, respectively. These wavefunctions were expanded in terms of the partial waves. In the direct amplitude f_d^r , we have denoted by \mathbf{r}_1 the position vector of the incident and scattered particle, while \mathbf{r}_2 stands for the position vector of the bound and ejected electron, respectively. In f_{ex}^r the states corresponding to the scattered and ejected particles were exchanged. ϕ_b^r denotes the bound state of the active electron. This molecular orbital has been constructed as a linear combination of Gaussian-type atomic orbitals, obtained as contractions of Gaussian-type functions as described in [11] and [12]. For linear molecules, the ϕ_b^r molecular orbital was expanded in terms of the Legendre polynomials

$$\phi_b^r(\mathbf{r}_2) = \sum_{l_b} C_{l_b}^r(r_2, R) P_{l_b}(\cos \omega_2), \quad (5)$$

while in the case of nonlinear molecules the expansion was performed in terms of the spherical harmonics

$$\phi_b^r(\mathbf{r}_2) = \sum_{l_b \mu} C_{l_b \mu}^r(r_2) Y_{l_b \mu}(\hat{\mathbf{r}}_2). \quad (6)$$

Here, $R = |\mathbf{R}|$ is the internuclear separation, while ω_2 stands for the angle between \mathbf{r}_2 and \mathbf{R} . The expansion coefficients in the above equations may be expressed as

$$C_{l_b}^r = \frac{2l_b + 1}{2} \int_{-1}^1 d(\cos \omega_2) P_{l_b}(\cos \omega_2) \phi_b^r(\mathbf{r}_2) \quad (7)$$

and

$$C_{l_b \mu}^r = \int d\hat{\mathbf{r}}_2 Y_{l_b \mu}(\hat{\mathbf{r}}_2) \phi_b^r(\mathbf{r}_2). \quad (8)$$

In order to calculate the ionization cross section, one must integrate in Eq. (1) over the angles of the scattered and ejected electron as well as the energy of the ejected electron.

Previously, we have investigated the positron impact ionization of several molecules (N_2 , CO, CO_2 and CH_4) in the framework of the CPE, ES and TS models [16]. These models were adapted for the electron impact ionization in the case of N_2 , CO, O_2 molecules [17]. We found that the ES model provides the best agreement with the experimental data in both cases. The ES model uses distorted waves in order to describe the ejected electron and plane or Coulomb waves to represent the incident and scattered particles. Details on these models are given in [16,17]. Based on the ES model, the determined cross sections for positron and electron impact ionization are compared.

3. Results and discussion

We have calculated cross sections for the ionization of the N_2 , CO, O_2 and CH_4 molecules by positron and electron impact. Our results, obtained in the framework of the ES distorted-wave model, are presented in Figs. 1–4. For electron impact ionization, we present two type of cross sections: direct (the curves denoted as e_d^-) and total (i.e. direct plus exchange: e_t^-), while for positron projectiles we have calculated direct cross sections (curves with e^+). For both projectiles, the theoretical results are compared with the corresponding experimental data [1–5,8].

Fig. 1 shows our results for the ionization of molecular nitrogen by positron and electron impact. The results show a very good agreement with the experimental data of Bluhme et al. [1] for positron impact ionization. For impact energies lower than 500 eV, the

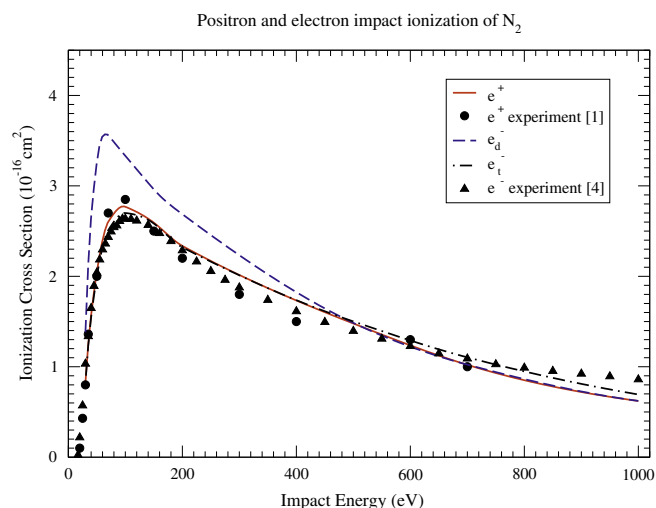


Fig. 1. Cross sections for the ionization of the N_2 molecule by positron and electron (direct and total) impact. The experimental points are from Bluhme et al. [1] and Straub et al. [4] for positron and electron impact, respectively.

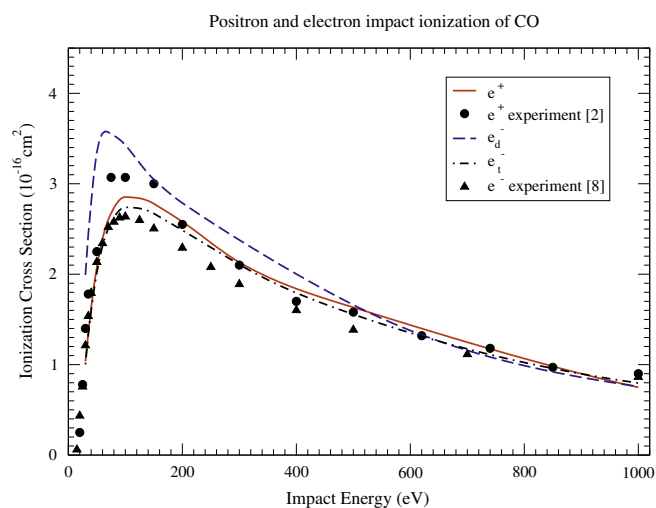


Fig. 2. The same as Fig. 1, but for the CO molecule. The experimental points are from Bluhme et al. [2] and Mangan et al. [8] for positron and electron impact, respectively.

cross sections for direct ionization by electron impact overestimates the experimental data of Straub et al. [4] and are higher than the cross section for direct ionization by positron impact. Taking into account the exchange effects, the cross section is diminished and agrees well with the experimental data. It can be observed that our calculated e^+ and e_t^- cross sections have similar behaviour like the experimental cross sections for positron and electron impact ionization. Particularly, in the region of the peak, the experimental data for positron projectiles are higher relative to the cross sections obtained in the case of electron projectiles, while for impact energies greater than 600 eV, the positron data seems to be a slightly lower. These features are reflected by our theoretical curves too.

For CO, the results are plotted in Fig. 2. We found a similar behaviour of the investigated cross sections as in the N_2 case. In the region of the cross section peak, the experimental positron cross section is higher than the electron cross section, while for energies greater than 600 eV, these two cross sections seems to merge. Excepting the energy range around the peak, where the e^+ cross section is a slightly lower than the experimental data, our

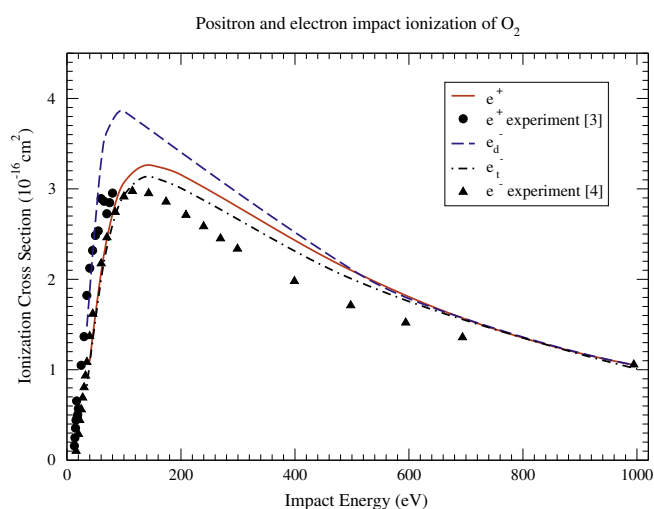


Fig. 3. The same as Fig. 1, but for the O_2 molecule. The experimental points are from Marler and Surko [3] and Straub et al. [4] for positron and electron impact, respectively.

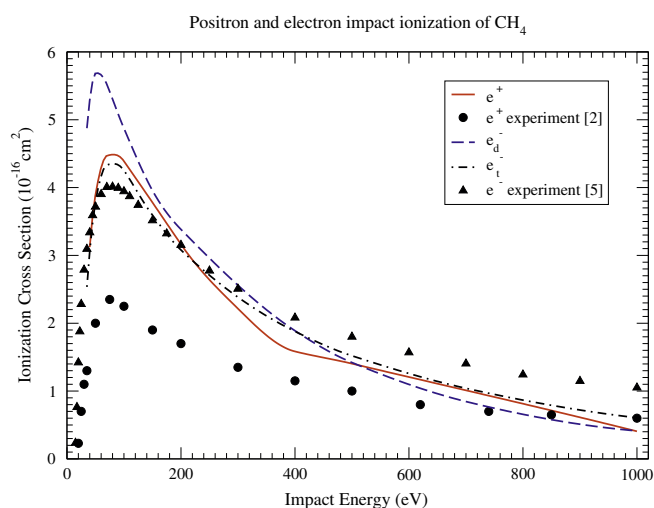


Fig. 4. The same as Fig. 1, but for the CH_4 molecule. The experimental points are from Bluhme et al. [2] and Straub et al. [5] for positron and electron impact, respectively.

results are in very good agreement with the measurements of Bluhme et al. [2]. For electron projectiles, mainly in the energy range from 100 eV to 600 eV, our e_t^- values are somewhat higher than the experimental results of Mangan et al. [8], while for higher energies we found a very good agreement. The theoretical electron cross section for direct ionization exceeds the corresponding positron cross section, mainly for energies below 500 eV. When the contribution from the exchange interaction is taken into account, the e_t^- cross section has lower values than the e^+ cross section. The difference between these cross sections is more significant at lower energies. It appears that for higher energies, the charge sign effect is insignificant.

In the case of the O_2 molecule, our results are presented in Fig. 3. The applied distorted-wave model produces a quite similar behaviour as found for the other two diatomic molecules. For energies falling below 500 eV, the direct electron cross section shows higher values than the direct positron cross section, while the total electron cross section is lower than the direct electron cross section for energies below 600 eV. The e_t^- curve lies below the e^+ cross

section values for the mentioned energy range. In the region of energies greater than 600 eV, the calculated theoretical cross sections are identical. The experimental data for the direct ionization of O_2 by positron impact are available only for impact energies lower than 80 eV [3]. It seems that the theoretical e^+ cross section lies somewhat below the experimental data of Marler and Surko [3]. Our results for electron impact ionization are in reliable agreement with the experimental cross sections for low and high impact energies. However, in the energy range from 100 eV to 800 eV the e_t^- cross section shows slightly higher values than the data of Straub et al. [4]. The larger discrepancy between the theoretical and experimental data found for this molecule, may be related to the more complex structure of the outer molecular orbital, where the oxygen molecule has two unpaired electrons.

The results for the CH_4 molecule are shown in Fig. 4. The calculated cross sections show similar arrangement for energies lower than 200 eV as in the case of the other studied molecules. For this energy range, the direct electron cross section has values above the positron cross section and the total electron cross section, while the latter lies below the positron cross section. For energies greater than 200 eV, the e_t^- cross section shows slightly higher values than the e^+ cross section. In the case of positron impact ionization our theoretical cross section overestimates the experimental results of Bluhme et al. [2], mainly for the energies around the peak, while for high impact energies the agreement was reliable. For electron impact ionization, our e_t^- results are in a good agreement with the experimental data below 60 eV and in the energy range from 160 eV to 400 eV. In the region of the peak our results are somewhat above the experimental data, while for energies higher than 400 eV the theoretical e_t^- curve lies slightly below the experimental measurements. It is interesting to notice that for this molecule, the experimental results for the electron impact ionization of Straub et al. [5] are much higher than the experimental data for positron impact ionization of Bluhme et al. [2], for the entire energy range, unlike for the other studied molecules. It may be that for this molecule we either do not understand a mechanism which makes the positron cross section much lower than the electron cross section and theoretical predictions, or there could be some problems with the absolute value of the experimental data.

4. Conclusions

We have performed DWBA calculations for the positron and electron impact ionization of the N_2 , CO, O_2 and CH_4 molecules. The calculated direct cross sections for electron impact ionization overestimates the experimental data, especially for lower impact energies. Below 500 eV, the direct electron cross sections are higher than the direct positron cross sections. By taking into account the exchange effects, the electron cross section was diminished. Our theoretical total (direct plus exchange) cross sections for electron impact ionization have lower values than the positron cross sections in the case of all studied molecules, mainly for impact energies below 300 eV. This is in accord with our expectations, since at lower energies the target molecule is polarized and the positively charged projectile meets a higher electron density relative to the electron projectile, which in turn leads to higher positron cross sections. Nevertheless, in the case of the CH_4 molecule, the experimental data of Bluhme et al. [2] for positron impact ionization are much lower than the data for electron impact ionization of Straub et al. [5]. Our theoretical cross sections for electron and positron impact ionization converge for high impact energies.

Our theoretical cross sections for electron impact are in good agreement with the experimental data, even in the case of the more complex CH_4 molecule, which suggests that the employed method may be applied to other complex molecules. However,

there is a discrepancy for the positron data, but at the moment we do not know its cause.

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