Electronic excitation of H_2, CO, and N_2 by positron impact

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In this paper we present results for positron impact excitation of electronic states of H_2 ($x^1\sum_g^+$ -> $B^1\sum_u^+$, $C^1\Pi u$), CO($X^1\sum_g \rightarrow a^1\Pi_g$), and, N_2($X^1\sum_g \rightarrow a^1\Pi_g$) using the scaling of plane wave Born cross sections. This method was originally proposed by Kim (Phys.Rev.A64032713(2001)) to electron scattering and adapted by Lino (*Chin. J. Phys.* **50** (2016) 223.) for positron scattering. Integral cross sections calculations are reported using this new method. For H_2 molecule our cross sections exhibit results comparable to the sophisticated theories, and experimental data. For isoelectronic molecules CO, and N_2 the results are compared to the *ab initio* Schwinger multichannel (SMC) method. For positron-CO scattering our model is very reasonable when compared with SMC method and as expected for positron-N_2 scattering, our results, as well as SMC method, failed to reproduce some structure observed by experimentalists. A important observation using the scaling plane wave Born for positron scattering is that the method is comparable to more sophisticated theories indicating the possibility of rapid and reliable calculations of excitation cross sections.

Keywords: Born; positron; scaling.

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1. Introduction

The interaction of positron with matter has long been a subject of intense research in the fields of both experimental and theoretical molecular physics [1]. New techniques and subsequent measurements of cross sections besides being directly in various other areas serve as a strong motivation for the theorists to extend their calculations to explore specific areas of molecular interaction with matter [1]. The comprehension of inelastic processes may be very useful in the near future to create alternative techniques to produce low energy positron beams and new theoretical studies. We can cite only few scientific publications on electronic excitation of molecules by positron impact. Among them, we point out a few studies as H_2 [2,3,4], N_2 [5,6,7], and CO [8,9]. With the advance of quantum mechanical computational methods, some very accurate ab initio calculations were performed and these calculations are very time consuming, limiting the domain of applicability of such models. In the last years, many analytical formulas have been developed to overcome these difficulties, some of them empirical and others derived from first principles [5].

Recently Lino [10] proposed a simple scaling of plane-wave Born cross sections for positron-impact excitation of atoms and molecules. The new method shown cross sections accuracy when comparable to reliable experimental data as well as to more sophisticated theories, such as the close coupling [11], and Schwinger multichannel (SMC) method[5,6]. Motivated by these results, the scaling of plane-wave Born was applied here to compute electronic excitation cross sections of positron by H_2 ($x^1\sum_g^+$ - > $B^1\sum_u^+$, $C^1\Pi u$ states), CO($X^1\sum_g \rightarrow a^1\Pi_g$), and, N_2($X^1\sum_g \rightarrow a^1\Pi_g$) molecules. The idea was to study the agreement between our integral excitation cross sections (ICS) with sophisticated theories. The focus of the present work is not the production of extremely accurate cross sections when compared with theoretical studies or experimental data but verify if our scaling Born method is capable, for instance, of describing similar agreement with theoretical studies or verify if the method is also useful to establish benchmark calculations at the Born level of approximation, indispensable in the development of a more complete formalism.

In Sec. 2 we identify the new method for positron scattering. In Sec. 3 we discuss the application of the method for e^+ - H_2, CO, and N_2 scattering. Conclusions are presented in Sec. 4.

2. Theory

The first Born approximation (FBA) is used as the starting point in the present work because (a) plane wave is the correct wave function at infinity for an positron (or electron) colliding with a neutral atom (or molecule), and (b) it is the simplest collision theory that uses target wave functions explicitly. The explicit use of target wave functions enables one to use relativistic wave functions for heavy atoms, and to distinguish the final state of the target. The scaling plane wave Born approximation described by Kim [12] for excitation of neutral atoms(molecules) is applicable to dipole-allowed excitations, and use some properties as excitation energy, ionization energy, and the dipole f value that can be obtained, in principle, from accurate wave functions. Since scaled cross sections are based on the plane wave Born approximation, they do not account for the resonances often found near the excitation thresholds. Qualitatively the FBA does not account for the electron scattering exchange effect with the target electrons, the distortion of plane waves in the vicinity of the target molecular, or the polarization of the target due to the presence of the incident particle. The scaling method [12]

combine these deficiencies into simple functional forms that depend on a few atomic or molecular properties. Cross sections for positron and electron impact are virtually identical at high energies and if the basic dynamical ingredients for this evidence is the FBA, then is possible extend the analysis developed by Kim [12] using more complicated systems, as for example, positron-molecule scattering. The modified scaling, to be referred to as the scaling Born positron (SBP), uses the excitation energy (E_{exc}), and positronium energy (E_{ps}) of the target and is valid only for dipole-allowed excitations. As showed in earlier study [10] the method not only reduces the cross sections magnitude at low energy, but also shifts the peak to a high energy than the peak of the unscaled FBA, while keeping the high energy validity of the FBA intact. In a generic form, FBA cross sections σ_{Born} (E) for inelastic collisions are written as

$$\sigma_{\text{Born}}(E) = 4\left(\frac{\pi}{E}\right) R a_0^2 F_{\text{Born}}(E) \tag{1}$$

where a_0 is the Bohr radius, R is the Rydberg energy, E is the incident positron energy, and $F_{\text{Born}}(E)$ is the collision strength (multiplied by a constant to be consistent with the standard definition of the collision strength). The scaling method apply only to integrated excitation cross sections, not to angular distributions, because the scaling method do not alter the angular distribution shape described by the unscaled Born cross sections. The modified scaling here proposed, replaces the E that appears in the denominator of Eq. (1), *i.e.*, E by $E + E_{\text{ps}} + E_{\text{exc}}$. The SBP method is introduced as

$$\sigma_{SBP} = \left[\left(\frac{E}{E + (E_{\text{ps}} + E_{\text{exc}})}\right)\right] \sigma_{\text{Born}}(E) \tag{2}$$

with the argument that the effective kinetic energy of the incident positron seen by the target is E plus the energy of the bound electron. Since the SBP method just an adaptation of the FBA, the cross section obtained gives a direct and unequivocal measure of the quality of the present model. A hint to the meaning of adding $E_{\text{ps}} + E_{\text{exc}}$ to E may be found in FBA cross section for the elastic scattering from the Yukawa potential. The potential is a screened Coulomb potential

$$V(r) = -\frac{z}{r} e^{(-r/\beta)} \tag{3}$$

where r is the radial coordinate, Z is the atomic number, and β is the "range" of interaction with a dimension of length. Using plane waves for the incident electron (or positron), the integrated cross section for elastic scattering is

$$\sigma_{el} = 16\pi Z^2 \beta^4/(1 + 4k^2\beta^2) \tag{4}$$

where k is the momentum of the incident positron in atomic units. After writing $\beta = b$ and noting that $k = E/R$ (Rydberg), Eq. (4) becomes

$$\sigma_{el} = 16\pi R Z^2 b^2 \beta^4/(E + R/4b^2) \tag{5}$$

which has the E in the denominator shifted by a constant with the dimension of energy. Although this analogy is not rigorous, the similarity between the Eq. (2) and Eq. (5) suggests that the constant ($E_{\text{ps}} + E_{\text{exc}}$) can be seen as the scaling factor to represent the correlation between the positron and electron of the target (the combination $E_{\text{ps}} + E_{\text{exc}}$ should not be taken literally as a rigid rule, but only as an indicator of the order of magnitude for the cross section) [10]. At present, the SBP cannot be "derived" from first principles but offers a simple and effective way to calculate cross sections comparable to sophisticate theories.

3. Results and discussion

The wave functions for the excited electronic states were generated for all targets using the improved virtual orbital (IVO) method [13]. For all targets the calculations were performed with the framework of the fixed-nuclei approximation at the equilibrium internuclear distance. The experience with some targets has shown that electronic excitation cross sections demand trial scattering basis sets with a higher degree of refinement than for pure elastic calculation. To illustrate, we present results of applications of the present method for inelastic scattering of positrons by H_2, CO, and N_2, for several impact energies. For all molecules we used Hartree-Fock calculations to represent the ground state of the targets with the same Cartesian Gaussian basis set expansion that have been used in previous calculations with the SMC method (Ref. 3 for H_2, Ref. 9 for CO, and Refs. 5 and 6 for N_2). Table I shows vertical excitation energies compared to experimental data for H_2 ($x^1\sum_g^+$ -> $B^1\sum_u^+$, $C^1\Pi u$), CO($X^1\sum_g \rightarrow a^1\Pi_g$), and, N_2($X^1\sum_g \rightarrow a^1\Pi_g$).

A.H_2

Recently, Lima and collaborators performed a series of studies on the electronic excitation of H_2 in the energy range from 13.6 to 30 eV using the SMC method [3]. The SMC method, by now, is considered to be a standard many-body approach for theoretical studies on the low-energy scattering of positrons by molecular target. A systematic set of procedures using the SMC method was used to verify e^+ - H_2 ($x^1\sum_g^+$ -> $B^1\sum_u^+$, $C^1\Pi u$) states. When dealing with dipole allowed transitions, the long-range character of the dipolar coupling requires a large number of partial waves and higher partial waves are not well described for the SMC method. To repair this problem, a FBA scheme combined with SMC method (called Born + SMC) was used in Ref. 3. For the

TABLE I. Excitation energies for H_2, CO, and N_2

Target	IVO	Expt
CO	8.07	8.07
N_2	10.34	8.55
H_2(B)	12.73	11.19
H_2(C)	13.12	12.30

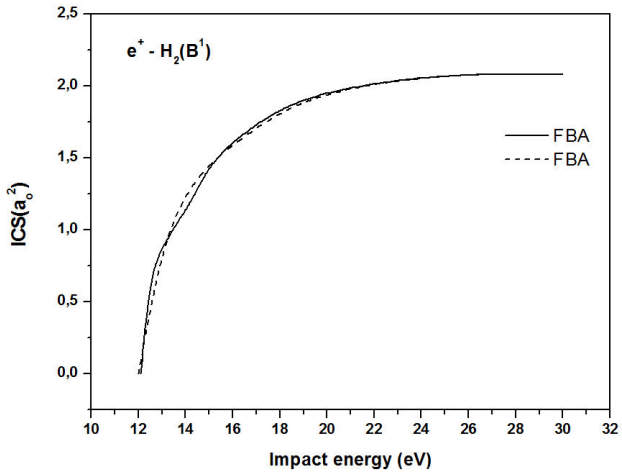

FIGURE 1. Electronic excitation cross section for H_2 ($X^1\sum_g^+$ -> $B^1\sum_u^+$) in units of a_o^2. Solid line: our FBA; dashed line: FBA of Lima *et al* [3].

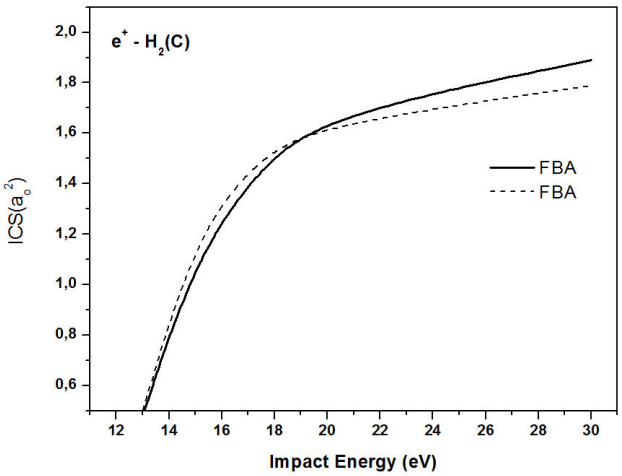

FIGURE 2. Electronic excitation cross section for H_2 ($X^1\sum_g^+$ -> $C^1\sum_u^+$) in units of a_o^2. Solid line: our FBA; dashed line: FBA of Lima *et al* [3].

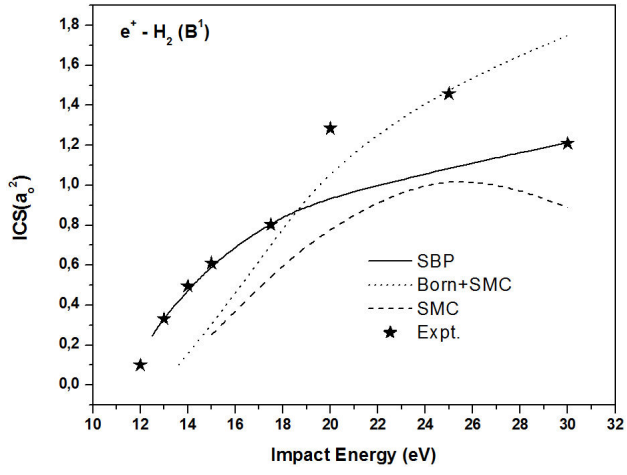

FIGURE 3. Electronic excitation cross section for H_2 ($X^1\sum_g^+$ -> $C^1\sum_u^+$) in units of a_o^2. Solid line: our SBP method; dashed line: SMC [3]: dot line: BSMC [3]: black star: experimental data [14].

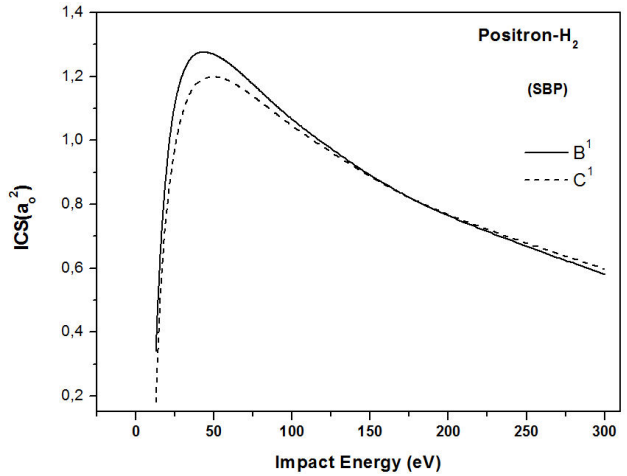

FIGURE 4. Electronic excitation cross section for H_2 ($X^1\sum_g^+$ -> $B^1\sum_u^+$, C^1) in units of ao2. Solid line, and dashed line: SBP method.

positronium formation threshold (E_{ps}) we have used 8.6 eV. Figure 1 show our FBA integral cross section for the $X\rightarrow B^1$ compared with FBA of Ref. 3 using the same basis set. As observed the results are satisfactory.

In Fig. 2 our FBA integral excitation cross section for the $X\rightarrow C^1$ is compared with FBA of Ref. 3 using the same basis set. As observed the results again are satisfactory.

In Fig. 3, we compare the calculated cross sections using SBP with SMC method [3], Born+SMC [3], and experimental data [14]. Analysis of the figure shows similarity of the SBP method with the SMC, and Born+SMC [3]. Note that our results are reasonable with experimental data [14] and is important to point out that this level of similarity is of special relevance, since the SMC method [3] demands great computational effort compared to the SBP.

In Fig. 4 we see reasonable convergence of the SBP method for B^1, and C^1 states. This figure also indicates a nontrivial result, *i.e.*, the cross section associated with $X\rightarrow C^1$ transition is very similar to the $X\rightarrow B^1$ one. This also was observed by Arretche and Lima using the SMC method [3]. These results can motivate experiments for the $X\rightarrow C^1$ transition.

B.CO

The second test problem chosen for the SBP method is the inelastic scattering from CO. Our study includes the $X^1\sum_g \rightarrow a^1\Pi_g$ electronic transition for impact energies up to 30 eV. For the positronium formation threshold (Eps) we have used 7.21 eV. In Fig. 5, our SBP cross sections for $X^1\sum_g \rightarrow a^1\Pi_g$ are compared to SMC method [9], Born+SMC method [9], and experimental data [9]. As observed in Fig. 5 the SBP method cross sections provide similar results (in shape) with the sophisticate SMC method [9], and as is well known in literature, the FBA approximation is larger in magnitude. It could be claimed that the SMC

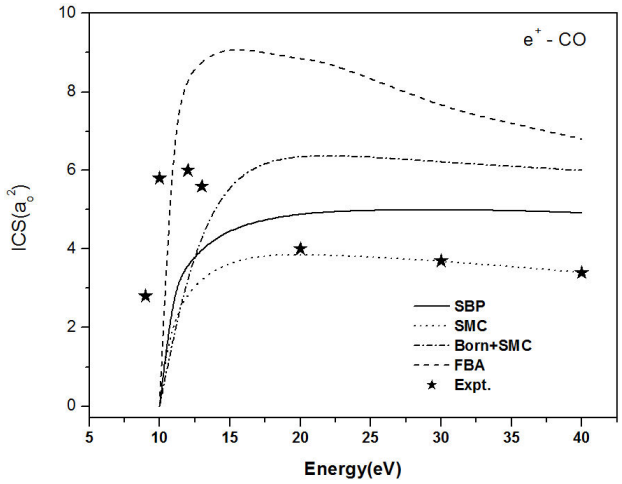

FIGURE 5. Electronic excitation cross section for CO($X^1\sum_g \rightarrow a^1\Pi_g$),, in units of a_o^2. Solid line, SBP method: Dashed line, FBA: dot line, SMC [9]: dot line, Born+SMC [9]: black star, experimental data [9].

method [9] and SBP calculations do not take nuclear motion. Results using the SBP method clearly represent a effective development theoretical and with significant contribution for positron-CO scattering.

C.N_2

The present calculation positron-N_2 scattering results are reported and compared with available theoretical study as the SMC method [6], and experimental data [16]. For the positronium formation threshold (E_{ps}) we have used 8.78 eV. The excitation cross section obtained with our FBA result is shown in Fig. 6. As observed in Fig. 6 our FBA is very similar with FBA used in Ref. 5.

In Fig. 7 we present cross sections for N_2 ($x^1\sum_g^+$ -> $a^1\Pi_g$) using our SBP method and SMC method [5,6] (SMC

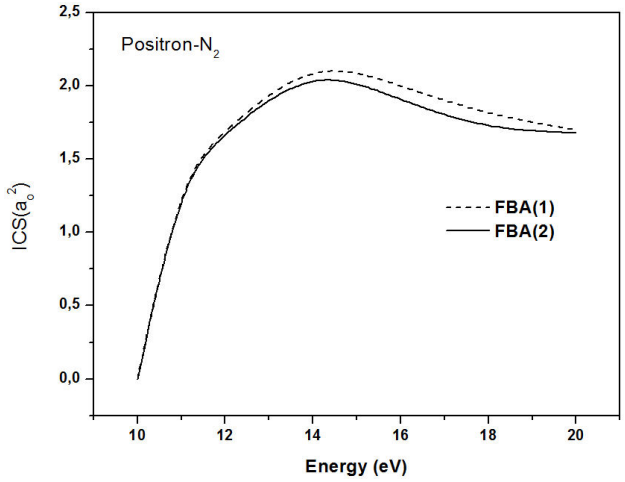

FIGURE 6. Integral cross sections(ICS) for e^+ - N_2 scattering considering the $X^1\sum_g \rightarrow a^1\Pi_g$ states. Solid line, our FBA(2); dashed line, FBA(1) of Ref. 5.

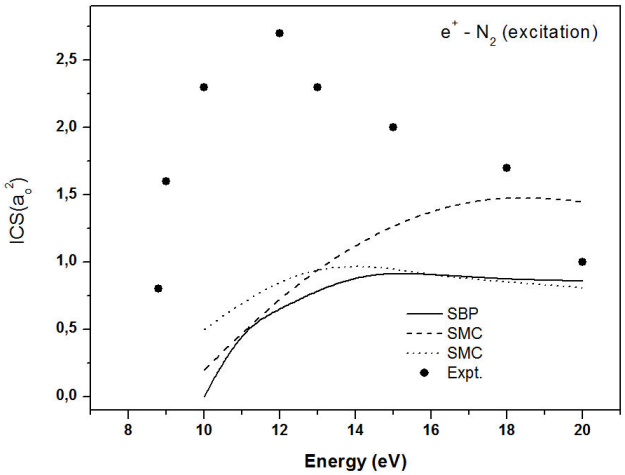

FIGURE 7. Integral cross sections (ICS) for positron-N_2 scattering considering the $X^1\sum_g \rightarrow a^1\Pi_g$ states. Solid line, SBP method; dashed line and dot line; SMC method using different basis set [5]; circle black, experimental data [15].

method using different choices for the basis sets). Theoretical results for this excitation failed to reproduce the near-threshold structure observed (measurements of absolute cross sections shown a striking near-threshold enhancement in the $a^1\Pi_g$ excitation cross section [15]). The SBP method proposed did not discuss the nature of the observed structure but the great similarity between our SBP and SMC method [5,6] (same order of magnitude) is noted. This fact is quite relevant because one of the most attractive features of the SBP is the possibility of well-describing cross sections comparable with sophisticate methods, for example, SMC [5]. Is evident that the SBP and SMC methods not reproduce the near-threshold structure observed in the experimental data [15] but is important mentioned that the SMC method [5,6] use a technique promising to separate unphysical (spurious) resonances from physical and this technique was designed to reproduce also the FBA [5,6]. As observed our SBP is more than reasonable and as cited before, is relatively simple. Nevertheless where such calculations are impractical or unavailable at the time, we believe that the SBP procedure does offer a very useful alternative.

4. Conclusions

The results presented in this paper demonstrate the utility of the scaling plane-wave Born approximation to obtain reliable positron-impact cross sections for H_2, CO, and N_2 molecules. This new method for positron scattering not only change the peak value, but also the cross sections shape at low energies. The method proposed can be used to predict cross sections, because the values of E_{ps} and E_{exc} are either available in the literature, or can be calculate from high quality wave functions, which can also be used to calculate the required FBA cross sections. The examples H_2, CO, and N_2 indicate that the scaling plane-wave Born is much better than using unscaled FBA cross sections. The results using the new method

not diminish the value of more sophisticate methods, though they require orders of magnitude more computational effort than SBP cross sections. The present scaling should also work, with minor adjustments if needed, on targets in general when Born cross sections have been calculated. Finally, applicability of the present scaling to molecular excitation cross sections should be a worth while topic to study and will facilitate the calculation of integrated excitation for several targets which pose difficulties to more computer intensive theories. The present SBP method is relatively simple compared to state-of-the-art ab initio theories.

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