

Positronium collisions with atoms and molecules

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Abstract. We describe recent theoretical efforts to explain the observed similarity between electron and positronium (Ps) scattering with both atoms and molecules. In order to perform these calculations at a more ab initio level we have developed a Free Electron Gas (FEG) approximation to the exchange and correlation potentials similar to that used in electron-molecule scattering. The FEG results give good agreement with experiment above the Ps ionization threshold and exhibit resonance features below the threshold. To take account of the effect of orthogonality between the scattered electron and the target wavefunctions we have used an orthogonalizing pseudopotential (OPP). The FEG plus OPP results give good agreement with experiment for Ps-rare-gas scattering. Preliminary results for Ps-H₂ scattering also agree well with experiment and demonstrate the importance of including orthogonality for scattering below the Ps ionization threshold.

1. Introduction

The interaction between positronium (Ps) and atoms, molecules and solids is important in such practical applications as positron annihilation spectroscopy [1] and positron emission tomography (PET) [2]. In both applications the production of gamma rays due to annihilation of Ps is influenced by its local environment. A better theoretical understanding of the effect of these interactions may help with the interpretation of these applications.

The theoretical study of Ps scattering is also important for understanding the results of recent experiments in which a beam of Ps is scattered by various atomic and molecular targets [3-5]. These experiments have shown that when the total electron and Ps scattering cross sections are plotted as a function of projectile velocity they are very similar in shape and magnitude above the Ps ionization (fragmentation) threshold which occurs at a Ps velocity of 0.5 a.u. (6.8 eV). This similarity is somewhat surprising since it seems to suggest that the positron is not as important as the electron in Ps collisions with atoms and molecules in this velocity region.

In electron collisions with atoms and molecules the static interaction is very important, although the exchange interaction can also have an important effect especially near resonances and thresholds. Correlation effects are also important in electron collisions due to the polarization interaction. In Ps collisions, on the other hand, the static and polarization interactions disappear due to the equal and opposite charges of the positron and electron in Ps. Long-range correlation effects are due to the weaker van der Waals forces between Ps and the target atom or molecule. This suggests that the exchange



interaction may be very important in Ps collisions. It also suggests that short-range correlation effects may be more important for Ps collisions with atoms and molecules than for electron collisions with the same targets.

The similarity between electron and Ps scattering above the Ps ionization threshold has been theoretically explained using the impulse approximation by Fabrikant and Gribakin [6] as mainly being due to the dominance of the exchange interaction in Ps-atom and Ps-molecule scattering. However, the impulse approximation is applicable only at high Ps velocities (energies). Therefore, other theoretical methods are needed for Ps velocities below the Ps ionization threshold. For these lower Ps velocities, the pseudopotential approach [7] has been developed. Both the impulse approximation and pseudopotential method use information on electron-atom and positron-atom scattering to develop a model for Ps-atom scattering. Specifically, in the pseudopotential method, a model potential (pseudopotential) is used in which the parameters of the potential are adjusted to reproduce the accurately calculated electron-atom and positron-atom phase shifts. The pseudopotential method also gives good agreement with experiment above the Ps ionization threshold [7,8] and again demonstrates the dominance of the exchange interaction in this region.

At velocities below the Ps ionization threshold the pseudopotential results for Ar, Kr and Xe exhibit a peak in the cross section [7,8]. This peak was explained [7] by a rapid decrease of the S-wave and the P-wave phase shifts in the low-velocity region. This behavior is very different from the results for the electron scattering cross sections for scattering from these atoms in this velocity region where a Ramsauer-Townsend minimum is observed.

On the other hand, the results of beam experiments that have recently been extended to velocities below the Ps ionization threshold [9] exhibit a very small cross section in this region which seems to suggest the possibility of a Ramsauer-Townsend minimum in Ps scattering with the heavy rare-gases. However, the pseudopotential method [7,8] as well as recent calculations by Green *et al.* [10] and Swann and Gribakin [11] have led to larger cross sections below the Ps ionization threshold for rare-gas atoms. It appears that there is some discrepancy between experiment and theory for Ps scattering by rare-gas atoms below the Ps ionization threshold. It should be noted that the beam experiments have thus far only been extended down to velocities of about 0.2 a.u. (1.1 eV) and only for the targets Ar and Xe. This means that there is a gap in the velocity (energy) range of about 0.06 a.u. (0.1 eV) - 0.2 a.u. (1.1 eV) for which practically no experimental data exists. Certainly, more experimental data is needed and there is a need for accurate theoretical calculations in this range.

Another interesting, but more complicated topic due to non-spherical interactions, is that of Ps-molecule collisions, for which the similarity between electron and Ps scattering has also been observed above the Ps ionization threshold in collisions with H₂, N₂, CO₂, O₂ and SF₆ [4, 5]. There have been relatively few theoretical studies of Ps-molecule collisions and these have been generally restricted to the simplest target H₂ [12-15].

We have previously applied the pseudopotential method to Ps collisions with H₂ by assuming that the Ps-molecule interaction is spherically symmetric [16]. While we obtained good results for H₂ in this way it turns out that severe difficulties are encountered when we attempt to use the pseudopotential approximation for more non-spherical systems like N₂. For this reason, we have recently developed a Free Electron Gas (FEG) approach to the calculation of exchange and correlation energies for Ps scattering [17]. From the target charge density, we can then use the Thomas-Fermi relation to obtain local exchange and correlation scattering potentials.

One deficiency of the local FEG potentials is that they cannot take into account the antisymmetric character of the total wave-function with respect to interchange of the projectile and target electrons. This deficiency can be rectified by enforcing orthogonality between the scattering wave-function and the bound orbitals of the target atom or molecule. Enforcing orthogonality has been shown to be important for electron-molecule scattering [18]. A method of enforcing orthogonality by using an orthogonalizing pseudopotential (OPP) for electron and Ps scattering has been proposed by Mitroy *et al.* [19,20]. An advantage of the OPP is that it does not require adjustable fitting parameters like the pseudopotential method of Refs. [7,8].

In the remainder of this report we describe our calculation of the local FEG potentials and nonlocal OPP. We then summarize our results of using these potentials to describe Ps scattering by rare-gas (Rg) atoms and several molecular targets.

2. Scattering potentials

A common method of including correlation in Ps scattering is to use the van der Waals potential with a short-range cutoff which is usually introduced in the form

$$V_{corr} = -\frac{C_W}{R^6} [1 - \exp[(R/R_c)^8]] \quad (1)$$

where C_W is the van der Waals constant, and R_c is an adjustable cut-off radius. In this way the long-range interaction is included properly, but the short-range correlation potential can depend significantly on the choice of cutoff radius [11].

In order to calculate the exchange and correlation effects in a more ab initio way we have computed the free-electron-gas exchange (FEG) and correlation energies, as a function of Fermi energy k_F using first and second order perturbation theory [17]. The energies obtained in this way can be converted to exchange and correlation potentials using the Thomas-Fermi model where the Fermi momentum depends on the charge density of the target atom or molecule through $k_F(\mathbf{r}) = [3\pi^2\rho(\mathbf{r})]^{1/3}$. The target charge density, $\rho(\mathbf{r})$, is calculated using accurate Hartree-Fock wave-functions. As described in Ref. [17] we evaluate the charge density at the Ps center of mass $\mathbf{R}$ so that $\mathbf{r} = \mathbf{R}$. The FEG potentials are similar to free electron gas approximations used in electron-molecule scattering [21,22]

One advantage of the current FEG correlation potential is that it can be matched smoothly to the long range van der Waals' form thus avoiding the necessity of an adjustable cutoff radius that affects the potential at small values of $\mathbf{R}$. We have found that the FEG correlation potential for Ps-rare-gas scattering is much more attractive at very small values of R than that of equation (1) [23].

For Ps-molecule scattering the charge density and local FEG potentials are not spherically symmetric. Therefore, in the case of molecules with Σ symmetry such as N_2 and CO_2 , we expand the FEG potentials in Legendre polynomials $P_\lambda(\cos \theta)$ depending on the angle θ between the molecular axis and $\mathbf{R}$. Also due to the molecular symmetry only even values of λ contribute to this expansion. For H_2 we have included terms up to $\lambda=12$ and for N_2 and CO_2 we have included terms up to $\lambda=24$. All of this is very similar to electron scattering from these molecules and we can describe the partial cross sections and eigenphase sums in a way similar to electron-molecule scattering, by conserved projection of the Ps angular momentum on the molecular axis and parity e.g. Σ_g , Π_u etc.

As mentioned above, we use the OPP to enforce orthogonality between the electron in Ps and the occupied target atomic or molecular orbitals, φ_i . The OPP is defined by the projection operator [19,20]

$$\gamma\hat{P} = \gamma\delta(\mathbf{r}_p - \mathbf{r}'_p) \sum_i^N |\varphi_i\rangle\langle\varphi_i| \quad (2)$$

where $\mathbf{r}_p$ is the positron coordinate. This operator is added to the exchange and correlation potentials and leads to a nonlocal term. The δ function ensures that the OPP affects only the electron coordinate, emphasizing in this way the dominant role of the electron constituent of Ps in the scattering. The strength parameter γ in these equations is made large enough so that orthogonality is enforced and the scattering calculations are converged in the sense that further increase of γ leads to a negligible change in the phase shifts and cross sections. For velocities below 2 a.u. the phase shifts for Ps-rare-gas scattering are converged for values of $\gamma=2$ a.u. and above and we have used a value of $\gamma=20$ a.u. in these calculations. For the present Ps- H_2 calculations we have used a value of $\gamma=3$ a.u. The sum in equation (2) is over the N occupied orbitals of the target atom or molecule.

We have derived an expression for the nonlocal OPP kernel describing Ps-Rg atom scattering in [23]. In the case of Ps-molecule scattering the situation is more complicated due to the non-spherical nature of the molecular orbitals. However, if we expand the molecular orbitals in Legendre polynomials and retain only the first, spherically symmetric term ($\lambda=0$) the expression for the kernel reduces to that of [23]. In our present calculations for Ps-H₂ scattering described in section 4 we make this approximation.

3. Ps-rare-gas scattering

As mentioned in the introduction the beam results for Ps-Rg scattering with Ar and Xe indicate the possibility that a Ramsauer-Townsend type minimum may occur in Ps scattering by the heavy rare-gas atoms, due to the sharp decrease in the cross section for velocities just below the Ps ionization threshold. The recent pseudopotential results [7,8] as well as the calculations of Swann and Gribakin [11], however exhibit larger cross sections below threshold with no Ramsauer-Townsend minimum.

Electron and Ps scattering might not be expected to be very similar in the low velocity region due to the fact that at these velocities the long-range interaction between the projectile and the target is very important, and this interaction is different for the two projectiles: for electron scattering the polarization potential decays as $1/r^4$, while for Ps scattering it is the significantly weaker van der Waals interaction which decays as $1/R^6$. The results of a number of experiments and calculations for Ps scattering by rare-gases [24] have resulted in a positive scattering length which seems to preclude the existence of a Ramsauer-Townsend minimum in Ps scattering by rare-gas atoms. This conclusion is further supported by the results of swarm experiments [24] which have also led to the conclusion that the Ps-rare-gas scattering length is positive. The swarm measurements are generally restricted to very low energies up to about 0.1 eV. Recently Shibuya and Saito [25] have developed a method to extract scattering phase shifts for Ps-Xe scattering from the experimental swarm data and, using effective range theory, have extrapolated their results up to an energy of about 1 eV. They obtain a peak with a very large magnitude in the Ps-Xe scattering cross section that seems to be somewhat similar to the peak obtained in the pseudopotential results. However, it seems that the use of the effective range theory over such a large velocity (energy) range is inappropriate and the huge magnitude of this peak seems physically unreasonable [23].

In order to help clarify the situation for low velocity Ps-rare-gas scattering we have recently applied the local FEG approximation and OPP described in section 2 to the calculation elastic Ps scattering with rare-gas (Rg) atoms [23]. The use of just the attractive, local FEG potentials leads to shape resonances at low Ps velocities that are not seen experimentally. The addition of the OPP to the local potentials removes the resonances and generally leads to good agreement with beam experiments [3,9,26], especially for the heavier atoms Ar, Kr and Xe. For He and Ne the cross sections are too large compared with experiment in the low-velocity region which may be due to the FEG potential not being as appropriate for such smaller systems since it is based on the statistical Thomas-Fermi model.

At velocities below the Ps ionization threshold the cross section is very sensitive to the intermediate and long-range correlation potential. This is because the exchange and correlation potentials are masked by the OPP at small values of R . In fact, for Xe we can get a Ramsauer-Townsend type minimum in the cross section if we use a potential of the form of equation (1) and adjust the cutoff parameter R_c so that the correlation potential is attractive enough in the region of intermediate R . This sensitivity illustrates the necessity of accurate determination of the exchange and correlation potentials in this region. The use of the local FEG exchange and correlation potentials with the OPP, in general leads to smaller cross sections and better agreement with results of beam measurements than previous pseudopotential calculations. We illustrate these results for Ar and Xe in figure 1. In these calculations we have included partial waves up to $l=10$. Note, that above the Ps ionization threshold previous calculations have shown that the excitation of Ps is negligible compared to elastic scattering and Ps ionization [6,27]. Therefore, we have added Ps ionization cross sections calculated by using the binary-encounter approximation [8] to the elastic cross section to obtain total cross sections in this region.

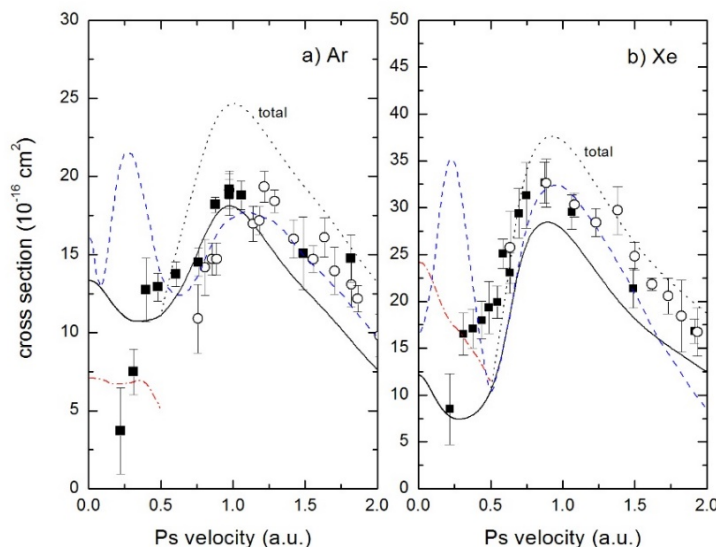


Figure 1. Cross sections for Ps scattering with a) argon and b) xenon. Solid line: elastic cross sections using the FEG potentials plus OPP of [23]. Dotted line: total (elastic plus Ps ionization) cross section. Blue dashed line: pseudo-potential results [8]. Red dash-dot line: results of [11] using the correlation potential of equation (1) with $R_c=2.5$ a.u. for Ar and $R_c=3.0$ a.u. for Xe. ■ [9]; ○ [3].

In the thermal energy region, inaccessible to beam experiments, our results compare well with swarm experiments measuring the Ps momentum transfer cross sections from observation of Ps moderation in gases. We obtain excellent agreement with measurements of Nagashima *et al.* [28] for He, Ne, and Ar, with Skalsey *et al.* [29] for Ne, and with Shibuya *et al.* [25,30] for Xe. However, the extrapolation of the Ps-Xe phase shifts, obtained by Shibuya and Saito [25] to higher energies leads to strongly overestimated (by an order of magnitude) total cross sections. As mentioned above this is due to the failure of the effective range expansion at higher energies.

It appears that our model provides a good description of Ps scattering by Rg atoms, especially the heavier atoms Ar, Kr and Xe. Our calculations confirm the positive sign of the scattering length and the absence of the Ramsauer-Townsend minimum in Ps scattering by heavier rare-gas atoms. However, the S- and P-wave scattering phase shifts decrease with energy significantly slower than in the previous pseudopotential calculations and significantly improves agreement with the beam experiments in the energy range below the Ps ionization threshold.

4. Ps-molecule scattering

The impulse approximation and pseudopotential method have also been applied with success to Ps collisions with molecular hydrogen [16]. In this work the Ps-H₂ interaction was taken to be spherically symmetric by retaining only the first term in the Legendre expansion of the e⁻-H₂ and e⁺-H₂ scattering potentials. For molecules with less spherical symmetry we have found that the pseudopotential method is difficult to implement since it is challenging to reproduce the scattering *T*-matrix elements using a model potential. Therefore, we have used the FEG model to construct local potentials and employed them in calculating scattering cross sections for Ps-N₂ collisions [31]. The resulting calculation is in good agreement with experimental measurements [32] above the Ps ionization threshold. In the present calculations we use partial waves up to $l=10$ for H₂ and $l=17$ for N₂ and CO₂.

An interesting feature of the experimental results is that there is evidence [4,32] for a resonance Ps-CO₂ state similar to the Π_u resonance in e⁻-CO₂ collisions, and for a Ps-N₂ resonance state similar to the

Π_g resonance in e^- -N₂ collisions near the Ps ionization threshold. Our recent results for Ps-N₂ scattering [31] also exhibit resonance feature in this region. The position of the resonance peak is slightly below the observed position which could be due to not including the effect of nuclear motion and/or orthogonality through the OPP. The effect of the nuclear motion on the Π_g resonance in electron scattering by N₂ is well known [33]. Therefore, a next step in the theoretical development should be incorporation of vibrational dynamics along the lines of the boomerang model [34]. This also opens an opportunity for the calculation of vibrational excitation cross sections in Ps-N₂ collisions. Below the Ps ionization threshold our cross sections for Ps-N₂ scattering [31] seem again to be larger than the experimental beam measurements [32]

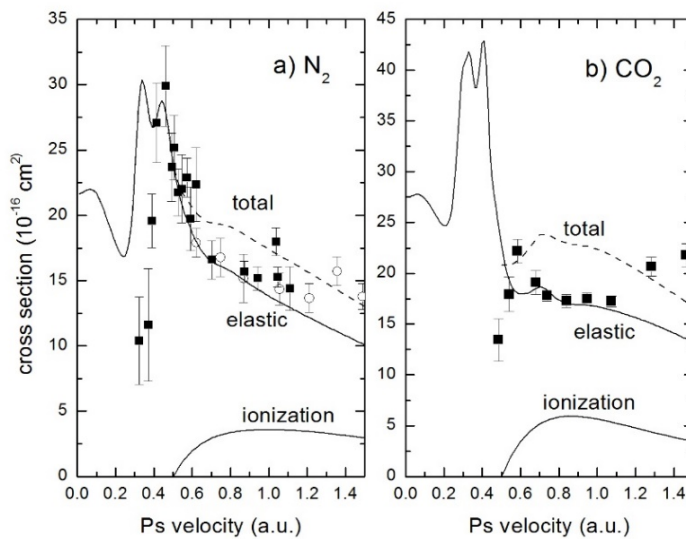


Figure 2. Cross sections for Ps scattering with a) N₂ of [31] and b) CO₂ using the FEG approximation. Solid lines: elastic cross sections. Dotted lines: total cross section (elastic plus Ps ionization). For N₂; ■ [31]; ○ [32]. For CO₂; ■ [4].

Calculations using the FEG exchange and correlation potentials for Ps-CO₂ scattering also show resonance features as demonstrated in figure 2. In this case the resonance position is further below the Ps ionization threshold and larger in magnitude. Above the Ps ionization threshold we add Ps ionization cross sections to the elastic FEG results using the binary-encounter approximation which was extended to Ps-molecule scattering in [31]. Our total cross sections in this region exhibit quite good agreement with the experimental measurements of [4]. From these results and those for N₂ it seems that the FEG model of the exchange and correlation potentials give a good description of Ps-molecule scattering above the Ps ionization threshold. Below the threshold we see resonance features, but the magnitude of the cross section is generally larger than the experimental beam results. We note that in our calculations of Ref. [31] we have used only the attractive local FEG potentials and have not included the repulsive OPP. Therefore, a next step for scattering below the Ps ionization threshold would be to develop the OPP for Ps-molecule collisions and see if it leads to better agreement with the experimental results in this region.

At this point we have made preliminary calculations in which the FEG plus OPP model is applied to Ps-H₂ scattering. In calculating the OPP in this case we have made the approximation of including only the first (spherically symmetric) term in the expansion of the occupied σ_g orbital. This approximation allows us to calculate the OPP in the same way that was done for Ps-He scattering in [23]. Although we have made the spherically symmetric approximation for the OPP, non-spherical terms have always been included in the calculation of the FEG potentials.

The results for Ps-H₂ in this case are shown in figure 3. Once again, we see a sharp resonance feature below the Ps ionization threshold when only the FEG potentials are used. When the OPP is added to the FEG potentials its repulsive nature suppresses the resonance and reduces the cross section below the threshold and gives good agreement with the measurements of Skalsey *et al.* [29,35] at low velocities and of Garner *et al.* [26] at velocities above the Ps ionization threshold. This is similar to Ps-Rg scattering and shows the importance of including the effect of orthogonality in Ps-molecule scattering. We are currently in the process of developing the OPP for Ps-N₂ and Ps-CO₂ collisions.

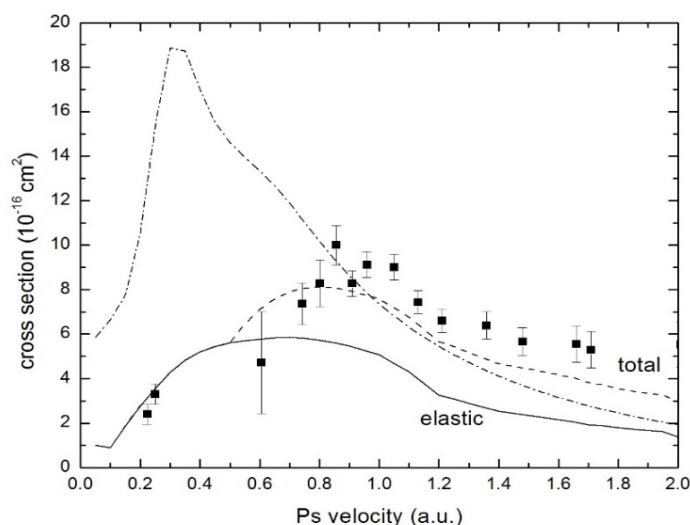


Figure 3. Cross sections for Ps scattering with H₂. Solid line: elastic cross section using the FEG potentials plus OPP. Dotted line: total cross section (elastic plus Ps ionization). Dash-dot line: Elastic cross section using the FEG approximation only. ■ Measurements of [29,35] (low velocities) and [26] ($v=0.6$ a.u. and above).

5. Conclusion and outlook

In summary, we have described our recent calculations of the total cross section for Ps scattering by rare-gas atoms and several molecules. In these calculations we have employed a FEG model for the calculation of exchange and correlation potentials. The FEG model gives good agreement with experimental results for the total cross section when we add Ps ionization cross sections calculated using the binary encounter approximation.

Below the Ps ionization threshold, the FEG results generally exhibit resonance behavior due to the attractive nature of the FEG potentials. For Ps-Rg scattering it is important to include the effect of orthogonality between the target and projectile electrons. We have developed the OPP to describe this effect. When the OPP is added to the FEG potentials we get relatively good agreement with experimental results for the heavy rare-gases, Ar, Kr and Xe, even at velocities below threshold. This illustrates the importance of including the effect of orthogonality at low velocities. Although we get better agreement with experiment, we do not see evidence for a Ramsauer-Townsend minimum.

For Ps-molecule scattering, when the FEG approximation is used, we again get good agreement with experiment above the Ps ionization threshold and see resonance structures near or below the threshold. For Ps-N₂ scattering the position and magnitude of resonance features agree quite well with experiment, but for Ps-CO₂ and Ps-H₂ collisions the resonance features have a large magnitude and appear at velocities significantly below the Ps ionization threshold.

We have begun the development of the OPP for Ps-molecule scattering by using a spherically symmetric approximation for the Ps-H₂ OPP kernel. When this is used along with the FEG potentials to describe Ps-H₂ scattering the resonance features are suppressed and the results agree well with experiment over the entire velocity range.

We are currently working on extending the OPP to more non-spherical molecules like N₂ and CO₂. Future work will also incorporate vibrational motion which will allow the calculation of vibrational excitation and other nuclear motion dependent processes.

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