S-wave elastic scattering of $\alpha$-Ps from $H_2$ at low energy

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The confined variational method is applied to investigate the low-energy elastic scattering of orthopositronium from $H_2$ by first-principles quantum mechanics. Describing the correlation effect with explicitly correlated Gaussians, we obtain accurate S-wave phase shifts and pick-off annihilation parameters for different incident momenta. By a least-squares fit of the data to the effective-range theory, we determine the S-wave scattering length, $A_s = 2.06a_0$, and the zero-energy value of the pick-off annihilation parameter, $Z_{eff} = 0.1858$. The obtained $Z_{eff}$ agrees well with the precise experimental value of $0.186(1)$ (J. Phys. B 16, 4065 (1983)) and the obtained $A_s$ agrees well with the value of $2.1(2)a_0$ estimated from the average experimental momentum-transfer cross section for Ps energy below 0.3 eV (J. Phys. B 36, 4191 (2003)).

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Introduction. Scattering of Positronium (Ps), i.e., a hydrogen-like atom composed of electron and positron, from atoms and molecules is fundamentally important for understanding the interaction between matter and antimatter [1–17]. Ps can be in a spin singlet state (para-positronium; $p$-Ps) or a spin triplet state (ortho-positronium; $\alpha$-Ps). Pick-off quenching is the process that the positron in the $\alpha$-Ps annihilates on collision with a molecular electron in the opposite spin state. The accuracy of experimental determination of the pick-off annihilation parameter $Z_{eff}$ of $\alpha$-Ps interaction with different targets such as $H_2$, CH$_4$, and CO$_2$ [1, 2, 13] is far ahead of that achieved by theoretical methods. The experimental results therefore can be used to test the quality and efficiency of theoretical methods, in particular the accuracy of the generated scattering wave functions. The complicated short-range electron-positron and electron-electron correlations as well as the electron exchange between Ps and target play key roles in the low-energy scattering of Ps. Theoretically, however, the accurate description of these interactions is very difficult and tedious due to the complex nature of a multi-centre scattering system.

In this Letter, we present confined variational studies of the low energy scattering properties of the experimentally studied $\alpha$-Ps-H$_2$ system. The work extends the \textit{ab-initio} theoretical description of the scattering of a composite projectile from a one-center target to a multi-center target. The obtained zero-energy value of the pick-off annihilation parameter, which is calculated for the first time ever, and the scattering length show excellent agreement with experiments [2, 8], demonstrating the high accuracy of the confined variational method (CVM).

The CVM [18–20] was first developed by Mitroy \textit{et al.} to accurately determine phase shifts of the low-energy elastic scattering of electrons ($e^-$) or positrons ($e^+$) from few-$e^-$ atoms in 2008. In 2012, the CVM was further developed by Zhang \textit{et al.} [20] to study the scattering of projectiles with internal structure, such as Ps. The CVM phase shifts for the S-wave $e^-$-He scattering at wave number $k = 0.2a_0^{-1}$ and for the S-wave Ps-H elastic scattering at $k = 0.1a_0^{-1}$ and $k = 0.2a_0^{-1}$ have set a benchmark for other theoretical methods [13]. In addition, the CVM was used to generate basis sets of energy-optimized explicitly correlated Gaussian (ECG) functions for other collision calculation methods such as the stabilization method [19] and Kohn variational method [21].

The remainder of this Letter is organized as follows. First, we briefly review the CVM using the $e^+$-potential scattering as an example. The reader is, however, referred to the papers [18, 21, 22] for a full account. Second, we numerically verify the CVM by calculating the phase shift and annihilation parameter of $e^+$ scattering from an H atom, giving also a comparison to other methods. Then the CVM is applied for studying the scattering of $\alpha$-Ps from H$_2$ at low energy.

Theory. Phase shifts are expressed in radians and atomic units are used throughout the following considerations, unless otherwise stated. Investigation of elastic scattering of $e^+$ from a short-range spherically symmetric
potential, which “represents” the short-range interaction between a positron and spherical many-electron atom, essentially means solving the Schrödinger equation

$$\left( -\nabla^2/2 + V_0(r) \right) \Psi_i(r) = E_i \Psi_i(r), \quad E_i > 0.$$ \hfill (1)

Assuming that the potential $V_0(r)$ is zero beyond a finite radius $R_0$, we may add a confining potential $V_{\text{CP}}(r)$ to the Hamiltonian in Eq. (1) in order to convert a complicated problem of many-body continuum states into much easier problems of many-body discrete bound states, one-dimension-potential bound states, and one-dimension-potential scattering. The Schrödinger equation of the confined many-body system becomes

$$\left( -\nabla^2/2 + V_{\text{aux}}(r) + V_{\text{CP}}(r) \right) \Phi_i'(r) = E_i \Phi_i'(r).$$ \hfill (2)

$V_{\text{CP}}(r)$ is typically chosen in the form \cite{13,22}

$$V_{\text{CP}}(r) = 0, \quad r < R_0,$$ \hfill (3)

$$V_{\text{CP}}(r) = G(r - R_0)^2, \quad r \geq R_0,$$ \hfill (4)

where $G$ is a positive number. Confining potentials of this type are chosen to avoid disturbing the $e^+$-potential interaction. Taking the discrete energies $E_i$ and expectation values $\langle \Phi_i'(r)|V_{\text{CP}}|\Phi_i'(r)\rangle$ as reference, we tune the auxiliary one-dimensional potential $V_{\text{aux}}(r)$ by solving the Schrödinger equation

$$\left( -\nabla^2/2 + V_{\text{aux}}(r) + V_{\text{CP}}(r) \right) \Phi_i'(r) = E_i \Phi_i'(r).$$ \hfill (5)

Like $V_0(r)$, $V_{\text{aux}}(r)$ has to satisfy the boundary condition $V_{\text{aux}}(r) = 0$ for $r \geq R_0$. The purpose of tuning $V_{\text{aux}}(r)$ is to achieve $E_i = E_i$ and $\langle \Phi_i'(r)|V_{\text{CP}}|\Phi_i'(r)\rangle = \langle \Phi_i'(r)|V_{\text{CP}}|\Phi_i'(r)\rangle$. To this aim, $V_{\text{aux}}(r)$ can be made flexible by inclusion of two or more parameters to adjust its shape and strength. For the elastic scattering of $e^+$ from an H atom, for example, $V_{\text{aux}}(r)$ is chosen in this work in the form

$$V_{\text{aux}}(r) = V_{\lambda_i,\alpha_i,\xi_i,\beta_i}(r) = \lambda_i(1 + 1/r) \exp(-\alpha_i r) + \xi_i r^2 \exp(-\beta_i r^2),$$ \hfill (6)

where $\lambda_i$, $\alpha_i$, $\xi_i$, and $\beta_i$ are the adjustable parameters. Equality of the energies means that the phase shift is the same and equality of $\langle V_{\text{CP}} \rangle$ ensures that the normalization condition at the boundaries is the same. Finally, the phase shift is obtained by solving the Schrödinger equation

$$\left( -\nabla^2/2 + V_{\text{aux}}(r) \right) \Phi_i(r) = E_i \Phi_i(r).$$ \hfill (7)

The key point of the CVM is that the logarithmic derivatives of the wave functions $\Psi_i(r)$, $\Psi_i'(r)$, $\Phi_i(r)$, and $\Phi_i'(r)$ are exactly the same for the same energy $E_i$ at $R_0$, i.e.,

$$\Gamma_{\Psi_i}(R_0) = \frac{1}{\Psi_i(R_0)} \frac{d\Psi_i}{dr} \bigg|_{R_0},$$ \hfill (8)

$$\Gamma_{\Phi_i}(R_0) = \Gamma_{\Phi_i}(R_0) = \Gamma_{\Phi_i}(R_0).$$ \hfill (9)

In addition, the phase shift is a function of $\Gamma_{\Psi_i}(R_0)$, i.e., $\delta_0(E_i) = \int \Gamma_{\Psi_i}(R_0)$. Therefore, the phase shift obtained from solving Eq. (7) equals that of $e^+ - V_0(r)$ scattering.

The calculation of the annihilation parameters $Z_{\text{eff}}$ for $e^+$ scattering and $Z_{\text{eff}}$ for $o$-Ps scattering depends on the normalization of $\Psi_i'(r)$ to the scattering boundary condition. For $e^+ - H$ scattering, for example, the procedure for determining $Z_{\text{eff}}$ is as follows. First, the expectation value of $\delta(r_{e^-} - r_{e^+})$ is computed with $\Psi_i'$,

$$\langle \delta(r_{e^-} - r_{e^+}) \rangle = \langle \Psi_i'(r_{e^-}, r_{e^+}) | \delta(r_{e^-} - r_{e^+}) | \Psi_i'(r_{e^-}, r_{e^+}) \rangle.$$ \hfill (10)

Second, the ratio of $\Phi_i'(r)$ and the continuum radial wave function at $R_0$ is computed. For S-wave scattering this is $A_k = \Phi_i'(R_0)/(\sqrt{4\pi} \sin(kR_0 + \delta_0))$. Then $Z_{\text{eff}}$ is defined as

$$Z_{\text{eff}}(k) = \frac{\langle \delta(r_{e^-} - r_{e^+}) \rangle}{A_k^2 k^2}.$$ \hfill (11)

Scattering of $e^+$ from an H atom. To demonstrate the accuracy of $A_k$ in the CVM, we calculate $\delta_0$, $A_k$, and $Z_{\text{eff}}$ for the S-wave $e^+ - H$ scattering at $k = 0.2 a_0^{-1}$, using two sets of basis functions: inner and outer. The inner basis functions are chosen as ECG functions,

$$\phi_k = \exp \left( -\frac{1}{2} \sum_{ij} b_{k,ij} r_i \cdot r_j \right).$$ \hfill (12)

They are optimized using the stochastic variational method \cite{23,24}. The outer basis functions are expressed in the form

$$\Psi_i = \psi_H(r_{e^-}) \exp \left( -\frac{1}{2} \alpha_i r_{e^+}^2 \right),$$ \hfill (13)

$$\psi_H(r_{e^-}) = \sum_j d_j \exp \left( -\frac{\mu_j r_{e^-}^2}{2} \right).$$ \hfill (14)

The wave function of the H ground state, $\psi_H(r_{e^-})$, is written as linear combination of 20 ECG functions with energy $E_H = -0.4999999999$ Hartree. Moreover, $\alpha_i$ is defined by the relation $\alpha_i = 18.6/1.45 i^{-1}$ for $1 \leq i \leq 40$. To take into account the polarization effect of H, $V_{\text{aux}}(r)$ additionally includes the polarization potential

$$V_{\text{pol}}(r) = -\frac{\alpha_d}{2r^2} \left( 1 - \exp(-r^6/r_0^6) \right),$$ \hfill (15)

with the static dipole polarizability $\alpha_d = 4.5$ a.u. and cut-off parameter $r_0 = 2.16 a_0$. 

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Table I. Convergence of the results for S-wave \(e^+\)-H scattering at \(k = 0.2a_0^{-1}\) as function of the number \(N\) of ECG functions. \(k\): wave number; \(\delta_0\): phase shift; \(Z_{eff}\): annihilation parameter.

| \(N\) | \(k (a_0^{-1})\) | \(\delta_0\) (rad) | \(Z_{eff}\) |
|------|----------------|-----------------|--------------|
| \(N_{inner}\) | | | |
| 200 | 0.20000185 | 0.187536 | 5.482 |
| 300 | 0.20000636 | 0.187630 | 5.536 |
| 400 | 0.20000011 | 0.187646 | 5.545 |
| 500 | 0.20000009 | 0.187648 | 5.554 |
| \(N_{inner} + N_{outer}\) | | | |
| 240 | 0.20000072 | 0.187608 | 5.480 |
| 340 | 0.20000121 | 0.187646 | 5.535 |
| 440 | 0.20000002 | 0.187653 | 5.545 |
| 540 | 0.20000000 | 0.187654 | 5.553 |

\(N\) and \(k\) are related to \(\kappa\) procedure makes \(\kappa\) outer basis. As a consequence, the convergence of \(\delta_0\) and \(Z_{eff}\). During the optimization, \(\delta_0\) be satisfied by tuning only \(\lambda\). The results for S-wave \(e^+\)-H scattering and \(e^-\) scattering are close to the value calculated with 300 ECG function (5.536), the CVM final result of \(Z_{eff}\) is 5.553 is more accurate than the results of COP and TM methods. It turns out that \(Z_{eff}\) increases monotonically with the number of ECG functions but converges slowly. We estimate that the exact value of \(Z_{eff}\) falls within the range from 5.554 to 5.559. We note that calculation with only a large inner basis has the capacity to generate accurate values for \(\delta_0\) and \(Z_{eff}\).

Scattering of \(o\)-Ps from \(H_2\). We employ the fixed nucleus approximation with an internuclear distance of \(R_{H_2} = 1.45a_0\), which is almost the equilibrium distance 1.448a0. Moreover, \(E_{H_2} = -1.174057 038\) Hartree as calculated by Rychlewski et al. [34] with 300 ECG functions is adopted for the ground state energy of \(H_2\). The Hamiltonian for the \(o\)-Ps-\(H_2\) scattering is

\[
H = -\sum_{i=1}^{4} \frac{\nabla_i^2}{2} + \sum_{j>i=1}^{4} \frac{q_i q_j}{|r_j - r_i|} \]

\[
+ \sum_{i=1}^{4} \left\{ \frac{q_i}{|r_i - \mathbf{R}/2|} + \frac{q_i}{|r_i + \mathbf{R}/2|} \right\},
\]

where \(r_i\) is the coordinate of the \(i\)-th particle \((e^+ or e^-)\) relative to the midpoint of the \(H_2\) molecular axis and \(q_i\) is its charge. The vectors \(\pm \mathbf{R}/2\) represent the displacements of the two protons from the midpoint. The basis for the interaction region has the form

\[
\phi_k = \hat{P} \exp \left( -\frac{1}{2} \sum_{i=1}^{4} b_{k,i} |r_i - \mathbf{S}_{k,i}|^2 \right)
\times \exp \left( -\frac{1}{2} \sum_{i=1}^{4} \sum_{j=i+1}^{4} a_{k,ij} |r_i - r_j|^2 \right).
\]

The vector \(\mathbf{S}_{k,i}\) displaces the center of the ECG function for the \(i\)-th particle to a point on the internuclear axis. The operator \(\hat{P}\) ensures that the basis has \(\Sigma_g\) symmetry. The confining potential is added in the center-of-mass region has the form

\[
V_{\lambda_{i},\alpha_{i}}(\rho) = \lambda_{i} \exp(-\alpha_{i}\rho - \frac{C_{\alpha}}{\rho}(1 - \exp(-\rho/\rho_0))
\]

with cut-off parameter \(\rho_0 = 6.0a_0\) and dispersion coefficient \(C_{\alpha} = 49.3\) a.u. [35]. Only the inner basis is used, because the outer basis is too complicated in this case. Similar to the case of \(e^+\)-H scattering, we expect that accurate scattering parameters can be obtained with a
large inner basis. In the following text, we use a superscript $T$ to indicate the triplet spin character of the pick-off annihilation. Due to the complexity of the multicenter scattering system, variational optimization of the energy and $\delta_T^{ep}$ together is very time consuming. Hence, only the energy is optimized by adjusting the nonlinear parameters of the ECG functions.

Table III addresses the convergence of the results for $\Sigma_g$ o-Ps-H$_2$ scattering at $k = 0.1a_0^{-1}$ as a function of the number $N$ of ECG functions. $k$: wave number; $\delta_T^{ep} = \langle \delta^2 (r_x - r_+) \rangle$; $\delta_0$: phase shift; $^{12}Z_{\text{eff}}$: pick-off annihilation parameter.

| $N$ | $k(a_0^{-1})$ | $\delta_T^{ep}$ | $\delta_0$ (rad) | $^{12}Z_{\text{eff}}$ |
|-----|----------------|-----------------|-----------------|----------------|
| 2400 | 0.100061       | 8.4043×10$^{-6}$ | −0.1876         | 0.1637         |
| 2800 | 0.100018       | 8.4635×10$^{-5}$ | −0.1863         | 0.1659         |
| 3200 | 0.100006       | 8.4687×10$^{-5}$ | −0.1859         | 0.1662         |
| 3600 | 0.100002       | 8.4873×10$^{-5}$ | −0.1857         | 0.1668         |

Table III presents results of our CVM calculations for $\Sigma_g$ o-Ps-H$_2$ scattering at $k = 0.1a_0$ when the number of ECG functions increases. We have $k = 2\sqrt{(E_1 - E_{Ps} - E_{H_2})}$, where $E_1$ is generated with the confining potential parameter $G = 1.7666 \times 10^{-4}$ (obtained from the optimization of the nonlinear parameters) and $E_{Ps} = -0.25$ Hartree is the exact energy of the Ps ground state. In Eq. (15), $\lambda_i$ and $\alpha_i$ have to be tuned together for each basis to satisfy the requirements to $k$ and $\langle \Phi_i (r) | V_C | \Phi_i (r) \rangle$. For a basis with 3600 ECG functions, for example, we obtain $\lambda_i \approx -0.382742$ and $\alpha_i \approx 0.553$. Both $k$ and $\delta_0$ show good convergence for an increasing number of ECG functions, in contrast to $\delta_T^{ep}$ and $^{12}Z_{\text{eff}}$ (though they vary monotonically).

Table III. Confining potential parameter $G$, $\delta_T^{ep} = \langle \delta^2 (r_x - r_+) \rangle$, phase shift $\delta_0$, and pick-off annihilation parameter $^{12}Z_{\text{eff}}$ for $\Sigma_g$ o-Ps-H$_2$ scattering at different $k$. Experimental values of $^{12}Z_{\text{eff}}$ are listed for comparison. Numbers in parentheses give the uncertainty in the last digit.

| $k$ $(a_0^{-1})$ | $G$ | $\delta_T^{ep}$ | $\delta_0$ (rad) | $^{12}Z_{\text{eff}}$ |
|----------------|----|----------------|-----------------|----------------|
| 0.06098       | 2.15×10$^{-6}$ | 1.7904×10$^{-6}$ | −0.1215         | 0.1737         |
| 0.08280       | 2.27×10$^{-6}$ | 4.7094×10$^{-5}$ | −0.1547         | 0.1687         |
| 0.10000       | 1.7666×10$^{-4}$ | 8.4873×10$^{-5}$ | −0.1857         | 0.1668         |
| 0.2           | Effective-range theory | 0.1858 |                     |                |

Exp. at 77.4 K [1] 0.197(3)
Exp. at 250 K [1] 0.195(5)
Exp. at 293 K [1] 0.193(5)
Exp. at 293 K [2] 0.186(1)

Table III presents results of our CVM calculations for three values of $k$. We focus our attention on scattering with $k \leq 0.1a_0$ for two reasons. First, the most reliable experimental information comes from annihilation experiments of thermal o-Ps. Second, the collision can be treated as S-wave scattering and, thus, the molecular aspects of the asymptotic wave function can be neglected. By fitting $\delta_0$ from Table III to the effective-range theory [30],

$$k \cot(\delta_k) = -\frac{1}{A_4} + \frac{1}{2} r_0 k^2 - \frac{4 \pi C_6}{15 A^2} k^3 - \frac{16 C_6}{15 A_s} k^4 \ln(k),$$

(19)

the scattering length $A_s = 2.06a_0$ and effective range $r_0 = 9.715a_0$ are obtained. The value of $A_s$ = (2.1 ± 0.2)$a_0$ estimated from the average experimental momentum-transfer cross section for Ps energy below 0.3 eV [8] agrees well with this result. The value of the pseudopotential method is much smaller (0.64$a_0$) [10]. In addition, our result for the S-wave cross section at $k = 0.1a_0$ (13.59$\pi a_0^2$) is much larger than the value of the three-Ps-state coupled-channel method (3.79$\pi a_0^2$) [3]. This means that both these methods seriously underestimate the near-zero-energy cross section.

Using the effective-range theory expansion [37],

$$^{12}Z_{\text{eff}}(k) = ^{12}Z_{\text{eff}}^{(0)} + ^{12}Z_{\text{eff}}^{(1)} k^2 + ^{12}Z_{\text{eff}}^{(2)} k^3,$$

(20)

fitting leads to $^{12}Z_{\text{eff}}^{(0)} = 0.186$. Experimental values of 0.197(3) [1], 0.195(5) [1], 0.193(5) [1], and 0.186(1) [2] from weighted least-squares fits of observed decay rates at low $H_2$ gas densities and temperatures of 77.4 K, 250 K, 293 K, and 293 K, respectively, indicate that the low-density $^{12}Z_{\text{eff}}^{(0)}$ is independent of the temperature (at the level of accuracy of the experimental data). The fit of Ref. [1] was constrained to a vacuum annihilation rate of $\Gamma_{\text{vac}} = 7.24 \mu$sec$^{-1}$, which is about 2.8% larger than the experimental value of 7.041(7) $\mu$sec$^{-1}$ [38]. Using no such constraint, a better value of $\Gamma_{\text{vac}} = 6.95(8)$ $\mu$sec$^{-1}$ was determined in Ref. [2]. We obtain perfect agreement with the experimental value of $^{12}Z_{\text{eff}}^{(0)} = 0.186(1)$ from Ref. [2].

Summary. The CVM is a powerful method that fully utilizes the advantages of studying bound states of atoms and molecules to determine phase shifts and normalization constants of asymptotic wave functions for collisions. The accuracy of the CVM normalization constant has been verified for $e^+\cdot H$ scattering by comparison with other methods. The CVM result of $Z_{\text{eff}} = 5.53$ for S-wave $e^+\cdot H$ scattering at $k = 0.2a_0^{-1}$ is the first significant improvement in accuracy since the COP value of $Z_{\text{eff}} = 5.538$ was reported in 1974 [27]. For o-Ps-H$_2$ scattering, we have reported accurate values of $\delta_0$ and $^{12}Z_{\text{eff}}$ for three different incident momenta. The CVM results for $^{12}Z_{\text{eff}}$ and $A_s$, extracted by means of the effective-range theory, show excellent agreement with precise experimental data [28]. As this problem was intractable for a long time, we believe that the present study will inspire new theoretical and experimental efforts on the low-energy o-Ps scattering from few-body targets.

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