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Spin polarization transfer in ground and metastable helium atom collisions

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Abstract. Cross sections and rate constants for total elastic, diffusion and transfer of metastability for collisions between He(1S)–He(3S) and He(1S)–He(3P) atoms over a wide range of energies and temperatures are presented. The rate constant for spin metastability excitation in He(1S)–He(3P) collision is several orders of magnitude larger than that for He(1S)–He(3S) collision at cold temperatures, due to attractive 3Πg potential interaction.

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1. Introduction

Spin-exchange optical pumping (SEOP) and metastability-exchange optical pumping (MEOP) are two techniques for producing hyperpolarized $^3$He nuclei with promising applications in science and medicine, including neutron spin-structure function [1], surface interactions [2], magnetic resonance imaging of lungs and internal organs [3, 4], fundamental symmetries [5] and NMR-based quantum computing, which rely on high nuclear spin polarization to achieve robust entanglement [6]. SEOP is accomplished by optical polarization of an alkali-metal atom electron spin, followed by collision transfer of spin polarization to $^3$He nuclei [7]. The collision is mediated by the isotropic Fermi-contact and anisotropic hyperfine interactions [8]. In MEOP, the nuclear polarization of the ground state $^3$He atoms accumulates in collision with optically pumped metastable triplet $^3$He atoms, in which the spin of the metastable electronic state is transferred to the former [9, 10]. The excitation transfer is mediated by the exchange of electrons, requiring the atoms to participate in close encounters.

Studies of the collision and spectroscopy of helium-containing systems are fundamental to understanding collisional ionization [11], Penning and associative ionization [12], spectral shift and broadening [13], loosely bound ultra-long-range helium dimers [14], Efimov trimers [15, 16], nanodroplets and bubbles in superfluid helium [17], high-precision measurements of fundamental constants [18] and degenerate quantum gases [19, 20]. Embedding of atomic and molecular species within finite helium nanodroplets, leading to the formation of metastable helium dimers in or on the helium nanodroplets, offers a sensitive probe of the physics of such finite quantum systems. Recently, He(23S) atoms were Bose condensed for the first time in a $^3$He buffer gas chamber [22] and were detected in time-of-flight imaging of the BEC cloud using the 1083 nm He(23S)–He(23P) transition. It is thus possible to investigate cold collisions of normal–metastable boson–fermion helium mixtures.

Helium atoms in their ground electronic state contribute negligibly to the chemistry of He-containing compounds: the He dimer binding energy is 1.3 mK [14]. The excited metastable state (He(23S)) lives for more than 8000 s [23] and contains a large reservoir of internal energy (about 20 eV). In the He(23P) state, helium lives for about 0.1 µs, thus behaving more like single active-electron alkali-metal atoms. Such a large reservoir of energy and the intricate behavior of the molecular potential energy curves in the metastable manifolds make large chemical reaction rates possible.

In this work, we investigate the collisional properties of normal and metastable helium atoms and obtain cross sections and rate constants for He(11S)–He(23S) and He(11S)–He(23P) collisions. The particular collision process that contributes to $^3$He nuclear polarization is the metastability transfer cross section ($\sigma_{tr}$). The quasimolecular state formed in a metastable–ground state He collision is a superposition of various states with different molecular symmetries. The cross section for transfer of metastability excitation is determined from scattering phase differences, associated with these superposition states, which differ significantly in the short range. We point out how the differing behaviors of molecular states in the He(11S)–He(23S) and He(11S)–He(23P) collisions produce dramatically different rate constants at very low energies.

Previous theoretical [24] results agreed qualitatively with older experimental data due to a poor representation of molecular interactions. The quality of the potential curves used in the present calculation is confirmed by comparison of their long-range behavior with analytical

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4 See the special topical issue on He droplets [21].
van der Waals (vdW) interactions, observed vibrational spectra [25, 26] and much better agreement with experiments.

2. Methods and results

We recently computed accurate molecular potential energy curves and long-range vdW coefficients. The collisional and radiative properties of the He(1S)–He(2S) system [25] and the pressure broadening and shift of the He(1S)–He(2P) line in the presence of ground state helium atoms [26] were calculated. These potential curves are used in the present work to investigate the collisional properties of the He(1S)–He(2S) and He(1S)–He(2P) system. Specifically, excitation transfer, diffusion and total cross sections are calculated for this system over a wide range of energies. These results could become useful in 3He buffer gas studies of metastability collisions.

2.1. Potential energy curves

The most accurate potential energy curves describing the ground–triplet state interaction in helium have been calculated [26] using a combination of the configuration interaction valence bond method and perturbation theory for a correct description of the long-range dispersive interaction [27]. The lowest six curves are displayed in figure 1. There is only one gerade–ungerade pair of molecular states that connects asymptotically to the He(1S)–He(2S) limit.

Figure 1. Born–Oppenheimer potential energy curves between ground state and triplet metastable states of the helium atom. The insets show the convergence of the long-range interactions towards their corresponding separated-atom limit.
and both of them, the $a$ and $c$ potentials, have a repulsive barrier at large separations and a deep well at shorter separations. The same can be said about the other $\Sigma$ pair, $d$ and $g$ curves, correlated to the He($1S$)–He($2P$) system. The energy barrier for these curves is even higher, at about 1 eV, requiring an extremely high temperature to activate these reaction paths. However, one of the states from the remaining $f–b$ $\Pi$ pair is barrierless and is strongly attractive. This state, $b_3\Pi_g$, predominantly influences collisions in the He($1S$)–He($2P$) manifold for energies smaller than the activation barrier in the other channels. The influence of the other states is seen through the intricate interference pattern observed in the cross sections.

2.2. Collisional phase shifts

Partial wave scattering phase shifts $\eta_\ell$ have been calculated for each of the six molecular potential curves of interest. A log-derivative method [25], with excellent stability and accuracy properties, is used to calculate the nuclear wave functions for each potential. The cross sections are calculated according to the following rules [28]:

- **He($1S$)–He($^3S$) total elastic**
  \[
  \sigma_t = \frac{4\pi}{k^2} \sum_\ell (2\ell + 1) \sin^2 \delta_\ell. 
  \]

- **He($1S$)–He($^3S$) excitation transfer**
  \[
  \sigma_{tr} = \frac{\pi}{k^2} \sum_\ell (2\ell + 1) \sin^2 (\eta_\ell(a) - \eta_\ell(c)).
  \]

- **He($1S$)–He($^3S$) diffusion**
  \[
  \sigma_d = \frac{4\pi}{k^2} \sum_\ell (\ell + 1) \sin^2 (\delta_{\ell+1} - \delta_\ell). 
  \]

  For all $^3S$ collisions, $\delta_\ell = \eta_\ell(c)$ for even $\ell$ and $\delta_\ell = \eta_\ell(a)$ for odd $\ell$.

- **He($1S$)–He($^3P$) total elastic**
  \[
  \sigma_t = \frac{4\pi}{3k^2} \sum_\ell (2\ell + 1) \left( \sin^2 \delta_\ell + 2 \sin^2 \delta'_\ell \right). 
  \]

- **He($1S$)–He($^3P$) excitation transfer**
  \[
  \sigma_{tr} = \frac{\pi}{3k^2} \sum_\ell (2\ell + 1) \left( \sin^2 (\eta_\ell(d) - \eta_\ell(x)) + 2 \sin^2 (\eta_\ell(b) - \eta_\ell(f)) \right). 
  \]

- **He($1S$)–He($^3P$) diffusion**
  \[
  \sigma_d = \frac{4\pi}{3k^2} \sum_\ell (\ell + 1) \left( \sin^2 (\delta_{\ell+1} - \delta_\ell) + 2 \sin^2 (\delta'_{\ell+1} - \delta'_\ell) \right). 
  \]

  For all $^3P$ collisions, $\delta_\ell = \eta_\ell(d)$, $\delta'_\ell = \eta_\ell(f)$ for even $\ell$ and $\delta_\ell = \eta_\ell(g)$, $\delta'_\ell = \eta_\ell(b)$ for odd $\ell$.
The molecular symmetries, from figure 1, are indicated in brackets to tag the different phase shifts. The diffusion cross sections, \( \sigma_d \), exclude small-angle scattering and, in this sense, differ from the total elastic cross sections, \( \sigma_t \). Because the \( \Sigma \) molecular potentials have repulsive barriers in both \( \text{He}(1^1\Sigma)\text{–He}(2^3\Sigma) \) and \( \text{He}(1^1\Sigma)\text{–He}(2^3\Pi) \) manifolds, they contribute more significantly to the diffusion of metastability. Similarly, the large repulsive barriers of the \( \Sigma \) potentials should suppress the transfer of metastability, reducing the transfer cross section, \( \sigma_{tr} \), especially at low energies—a fact first discussed in [29]. In the \( \text{He}(1^1\Sigma)\text{–He}(2^3\Pi) \) collision, on the other hand, the fully attractive \( b^3\Pi_g \) potential is instrumental in the transfer of metastability, producing large \( \sigma_{tr} \), and in particular at very low energies, where the attractive vdW interaction in the \( \Pi \) state dominates the collision. This is also true for the very low-energy diffusion of metastability.

2.3. Cross sections

The results of the present calculations are presented in figure 2. Due to interference among various scattering channels, the cross sections display a rich resonance structure at all energies for \( ^3\Pi \) collisions and only at large energy for \( ^3\Sigma \) collisions. This is due to the fact that collisions with \( ^3\Sigma \) atoms at low energy are impeded by the potential barrier in the \( a^3\Sigma_u^+ \) and \( c^3\Sigma_u^+ \) potential curves, and therefore the cross sections are dominated by the universal long-range part of the interaction potential. This is in contrast to the amplified structures in the \( ^3\Pi \) channels, which remain open at all energies and abound in low-energy resonances.

The results agree fairly well with the experimental data, which are mostly available for \( ^3\Sigma \) collisions only. Previous theoretical estimates [24] are clearly in disagreement with the experimental results, although they capture qualitatively the main features of the cross sections. This demonstrates that the quality of potential energy curves has a strong influence on the calculated cross sections, especially in the range of energies considered here. The only other experiment on \( \text{He}(1^1\Sigma)\text{–He}(2^3\Pi) \) collisional excitation [30] measured the fine-structure mixing of different \( ^3\Pi \) levels and found the cross section at room temperature to be \( 68 \pm 3 \text{ Å}^2 \). Our calculated value [26] for the \( 3P_{j=0 \rightarrow j=1,2} \) mixing transfer cross section is \( 75 \text{ Å}^2 \). Where temperature is used to report measured cross sections, we convert to energy as \( E = k_B T \), where \( k_B \) is the Boltzmann constant. Some measured cross sections were obtained from the rate constants by dividing the mean velocity at a given temperature.

The total scattering cross section shown in figure 2(a) is about \( 140 \text{ Å}^2 \) for \( ^3\Sigma \) collisions at room temperature, in good agreement with the measurements presented in [31] and [32]. At the same energies, the \( ^3\Pi \) total cross section is about \( 267 \text{ Å}^2 \), but it has a more pronounced decrease with increasing energy.

The suppression of metastability transfer for \( ^3\Sigma \) collisions, due to the energy barrier in both channels, is correctly described by the present model, as illustrated in the inset in figure 2(b). One channel is always open in \( ^3\Pi \) collisions; therefore, the cross section for excitation transfer is not suppressed and is about \( 80 \text{ Å}^2 \) at room temperature. At larger temperatures, both cross sections converge to a plateau at \( 20 \text{ Å}^2 \). We note that while the \( c^3\Sigma_u^+ \) potential curve in the calculations of [24] has a barrier, it is entirely repulsive, so that no short-range phase can accumulate. In the \( ^3\Pi\text{–}^3\Sigma \) manifold, all curves other than \( b^3\Pi_g \) are repulsive in [24] at all values of \( R \) and therefore produce quite different cross sections.

The excitation diffusion cross section has a dip down to \( 10 \text{ Å}^2 \) for \( ^3\Sigma \) collisions, and at \( T = 300 \text{ K} \) its value is \( 21.5 \text{ Å}^2 \), as shown in figure 2(c), while the cross section for \( ^3\Pi \) is approximately \( 91 \text{ Å}^2 \). Again, at larger temperatures, both cross sections go to \( 20 \text{ Å}^2 \).
Figure 2. Total elastic (a), excitation transfer (b) and excitation diffusion (c) cross sections for He–He\(\ast\)(2\(^3\)S) (red curves) and He–He\(\ast\)(2\(^3\)P) (blue curves) as a function of collision energy. Solid lines are the present calculation, dotted (‘+’) and dashed (‘◦’) lines are from previous theory [24], purple diamonds are experiments of [32], green squares in (a) are experiments of [31], green squares in (b) are experiments of [9] and green squares in (c) are the same experiments as reported in [24]. All experimental data refer to He–He\(\ast\)(2\(^3\)S) collisions. Arrows on the right-hand side indicate the results of semiclassical calculations.

2.4. Semiclassical description

The large-energy collisional behavior of the cross sections can be modeled using the semiclassical impact-parameter approximation. The semiclassical phase shift, as a function of energy \(E\) and impact parameter \(b\), results from comparing collision actions in the presence of
Figure 3. Total elastic, diffusion and transfer rate constants for collisions between the ground state helium atom and the metastable helium atom in triplet S and P states, as a function of temperature. Most of the oscillations in the cross sections in figure 2 are thermally averaged out.

the interaction $V(r)$ between atoms, and without it:

$$
\eta(E, b) = k \left( \int_{r_0}^{\infty} \sqrt{1 - V(r)/E - b^2/r^2} \, dr - \int_{b}^{\infty} \sqrt{1 - b^2/r^2} \, dr \right),
$$

where $r_0$ is the outer turning point and $k$ is the wave number for the collision. The semiclassical cross section is obtained by integrating over the impact parameter. These cross sections are valid for high energies, and the arrows on the right-hand side in figure 2 indicate the semiclassical results for collisions at $10^4$ cm$^{-1}$: the total cross section for $^3P$ collisions (blue arrow) is 136 Å$^2$, and that for $^3S$ collisions (red arrow) is 106 Å$^2$. The semiclassical approximation for excitation transfer and diffusion provides the same value as indicated in figures 2(b) and (c) by arrows: 24 Å$^2$ for $^3P$ collisions (blue arrow) and 16 Å$^2$ for $^3S$ collisions (red arrow). As expected for collisions of massive particles at large energies, the agreement of the semiclassical approximation with the rigorous quantum calculation is excellent.

2.5. Rate constants

In figure 3, we give the rate constants for total elastic, transfer and diffusion collisions among He(1S)–He(2S) and He(1S)–He(2P) partners. At very low energies, the Wigner threshold behavior becomes evident, as expected. The transfer rate constants for He(2S) and He(2P) have different behaviors at cold temperatures, because for the He(2S) case the transfer is
suppressed due to the strong repulsive interaction, while in the He(2\(^3\)P) case the \(^3\Pi_\text{g}\) attraction is more than sufficient to compensate for repulsion in other channels. The He(2\(^3\)P) transfer rate constant is, therefore, several orders of magnitude larger, with implications for spin polarization transfer at cold metastability collisions.

In summary, the refined potential curves afford much improved agreement with the available measurements as compared with previous theoretical efforts. Metastability transfer is shown to occur with large rate constants for the cold collision of He(1\(^1\)S)–He(2\(^3\)P) species. Experiments on the metastability transfer of spin polarization in \(^3\)He buffer-gas cooled magnetic traps could potentially be useful.

Acknowledgments

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