TWO-ELECTRON EXCITATION IN SLOW ION-ATOM COLLISIONS: DE91 006397

INTERFERENCE AMONG AUTOIONIZING STATES

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ABSTRACT

Structures found in the Auger electron energy distribution that are due to interferences resulting from collisions of H^+ ions with He atoms below 5 keV are theoretically studied within a close-coupling scheme based on molecular representation. The present study identifies the dominant interference-related contributors to the structures. These contributors are due to the interferences between (i) different autoionizing electronic states, (ii) incoming and outgoing parts of collisions on a single trajectory, (iii) different locations that have the same ejected electron energy on a single trajectory, and (iv) different trajectories.

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The interaction time for a collision system of $H^+ + He$ below 5 keV is in approximately in the same order as lifetimes of various doubly excited states of the He atom. Therefore, the doubly excited states produced may decay through the Auger process by ejecting a continuum electron while the projectile is still near. Hence, the ejected electron is influenced by the Coulomb interaction of the projectile. Consequently, its energy and angular distributions are broadened and shifted. This circumstance offers a rare opportunity to study various interferences in the ejected electron energy spectrum (EEES) that arise from the molecular and atomic natures of the colliding system and from interactions among particles or so-called post-collision interactions (PCI). In addition, a careful analysis of the EEES may provide information about the signature of "dynamical" electron correlation (electron correlation during the collision).

Numerous experimental studies have investigated two-electron excitation in collisions of multiply charged ions with He and H_2 targets, which dominantly undergo two-electron capture processes.¹ Some of these experimental studies claimed to observe evidence of an electron correlation effect for two-electron capture. A typical example of such measurements is one by Stolterfoht et al.,² who observed Coster-Kronig electrons [one electron in a low- n ($\cong 3$) manifold and another in a high- n (≥ 6) manifold] along with the usual KMM Auger electrons in $O^{6+} + He$ collisions at $E = 60$ keV. At the request of experimenters, several theoretical studies based on semiquantitative treatments have also appeared within approximately the last year, seeking to understand these experimental findings.³

In contrast, experimental studies using lowly charged ions such as H^+ or He^+ as projectiles are very rare because the two-electron process dominantly proceeds through excitation. Hence, the cross section for the process is very small at low-keV collision energies, making experiments extremely difficult. One recent experiment by van der Straten et al.⁴ dealing with the EEES from He doubly excited states that result from collisions with Li^+ ions in the energy range from 1 keV to 5 keV used a coincidence technique. The authors observed rich structures in the EEES and provided a semiempirical analysis that appears to

interpret the origin of the structures with reasonable success. In addition, they were able to extract from their measured data excitation probabilities of $\text{He}(2s^2, 2s2p, 2p^2)$ doubly excited states as functions of impact parameters and collision energies. Although some theoretical attempts⁵ have been made to understand these structures in the EEES on the basis of a perturbative approach, the perturbative treatment is clearly not adequate to deal with problems of this kind because, in principle, the interference is due to a multichannel effect, in which many channels simultaneously play an essential role. Hence, autoionizing states and discrete Rydberg states should be treated equally within a close coupling scheme.

We have used a molecularorbital (MO) expansion method to study the topic. A brief summary of our theoretical approach is the following: (i) The semiclassical MO expansion method was used with inclusion of molecular electron translation factors. (ii) Discrete molecular states including doubly excited states were obtained by a two-electron configuration interaction method modified by inclusion of a pseudopotential for representation of atomic cores, if necessary. (iii) Molecular continuum states were obtained as elastic electron scattering states from a diatomic molecule by solving the static-exchange Hartree-Fock equation. (iv) Using these discrete and continuum molecular wavefunctions, nonadiabatic couplings among discrete states and autoionization widths between doubly excited and continuum states were rigorously calculated. (v) Seven discrete states and five continuum states plus five doubly excited states were used in the close-coupling calculation as an initial test calculation.

As an initial attempt, we chose the $\text{H}^+ + \text{He}$ collision system for our test study to elucidate the fundamental physics of the problem, although no experimental result has been reported on the EEES for the system at low-keV collision energies. Figure 1 displays adiabatic potentials of the HHe^+ system. Several potential curves between the initial ($\text{H}^+ + \text{He}$) and ionization ($\text{H}^+ + \text{He}^+$) levels correspond to one-electron excitation or capture processes. The most significant features in the potentials are sharp avoided crossings among autoionizing states including doubly excited states and Penning states within a very narrow

energy band. This feature suggests that many Rosenthal-type transitions among autoionizing states may occur, leading to out-of-phase scattering amplitudes.

Inspection of the potentials in Fig. 1 caused other possible sources of interference to be proposed for examination. These sources are the interferences between (i) M_L states in a single $(n\ell, n'\ell')$ manifold, (ii) nearby autoionizing states (mentioned above), (iii) different atoms in a symmetric system, (iv) direct ionization and autoionization, (v) autoionization at incoming and outgoing parts of a collision, (vi) different paths, and (vii) different locations on a single path with the same ejected electron energy. The sources (v)-(vii) lead to interferences because the phase factors evolve differently in time, at different R . In addition, a report exists of experimental evidence⁶ for interaction between an electron ejected by the Penning process and the nuclear motion of a residual molecular ion. The interference was caused by an exchange of angular momentum in the low-eV collision regime. However, in the present study, we excluded this possibility because the energy region of our interest is far higher than that of the experiment.⁶ We also neglected (iii) because our test system is not symmetric. We examined the possible causes of the interferences (i)-(vii) and their relative importance.

Figure 2 shows the EEES obtained by fully accounting for (i)-(vii). The general features of the EEES are (i) the dominant peak at $\epsilon \approx 35$ eV that is due to atomic autoionization (AAI) of the He atom, (ii) the broad peak on the lower energy side that is due to molecular autoionization (MAI) from the HHe^+ quasimolecule, and (iii) the PCI effect and the Doppler shift on the lower energy side. Helium doubly excited states for $2s^2\ ^1S$, $2p^2\ ^1D$, and $2s2p\ ^1P$ are considered to be the prime channels for excitation. Their nominal energies are 33.2 eV, 35.30 eV, and 35.54 eV, respectively, and the lifetimes are known to be in the range of 4×10^{-15} sec, 9×10^{-15} sec., and 15×10^{-15} sec, respectively.⁷ Hence, at the higher energy end of the present study, a combination of $2s2p\ ^1P$ and $2p^2\ ^1D$ autoionizing states is primarily responsible for the AAI peak, while the $2s^2\ ^1S$ state with the PCI effect and the Doppler shift tailing from two levels above constitutes the broad MAI peak. As the energy

decreases, one would expect that the AAI peak decreases and merges with the MAI peak to form a broad peak with abundant structures. The transition probability, I_{fi} , for autoionization is described in a coherent sum of amplitudes, written in a simplified form as

$$I_{fi} \propto \left| \sum_L A_L(t) g_{PCI}(\epsilon_L, \epsilon, D) V_{ed} Y_L(\Omega) \right|^2, \quad (1)$$

here $A_L(t)$ describes the transition amplitude for forming a doubly excited state with the representative quantum state L of the He atom, and $g_{PCI}(\epsilon_L, \epsilon, D)$ represents the PCI effect that describes the energy shift of an ejected electron from the nominal energy ϵ_L to ϵ and the Doppler shift D caused by the moving doubly excited He atom. The term V_{ed} represents the coupling matrix element between the autoionizing state and the ionized state (the HHe^{2+} core + one free electron), and $Y_L(\Omega)$ denotes all angular components of the ejected electron. Equation (1) clearly predicts that the various types of interferences in the EEES for the present collision system originate in the sources summarized as (i)-(vii) above.

The structure around 35 eV is resulted by the interference of the amplitudes to $2p^2 \ ^1D$ and $2s2p \ ^1P$ states and that seen at 33 eV is due primarily to $2s2 \ 1g$ state with some interferences from above states. The small structures seen between and on the AAI and MAI peaks are due to the causes discussed above. These causes were found to be more sensitive to the autoionization collision energy and impact parameter than to the collision energy and impact parameter for discrete transitions. However, at the lower end of the energy studied (0.5 keV), sources (ii), (vi), and (vii) appear to be dominant contributors to the structures in the EEES.

The present results, based on the calculation with a small basis set, seem to be promising. More systematic studies on the topic using a larger basis set are underway and will be reported elsewhere.

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Figure Captions

Fig. 1. Adiabatic potentials for the HHe^+ system. Levels below the ionization channel $[\text{H}^+ + \text{He}^+]$ belong to single-excitation/capture channels.

Fig. 2. The EEES obtained by the present calculation with $E = 0.8 \text{ keV}$ and an electron angle to the projectile of 120° . See the text for an interpretation of the structures.

Fig.1

Fig.2

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