Correlated electron detachment in H^--He collisions

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Experimental and theoretical studies of the energy spectra of electrons ejected in H^--He collisions have been performed. Comparison of calculations using the Sturmian and advanced adiabatic theories with experimental data reveals the existence of a correlated electron detachment mechanism, which yields the main contribution to formation of the high-energy part of the ejected electron energy distribution. This mechanism is associated with dynamical energy transfer to the loosely bound $1s'$ electron of H^- in the course of superpromotion of the inner $1s$ electron.

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I. INTRODUCTION

Studies of the dynamics of electron detachment of negative ions are of interest from both the fundamental and applied points of view. Fundamentally, it is important that the outer, weakly bound electron of a negative ion is attached to a neutral atom via non-Coulomb interaction, such that there is no Rydberg series and it is possible to study direct coupling of a single discrete state with the continuum. From the applied point of view, the behavior of negative ions, especially H^-, colliding with gas atoms and molecules is of considerable interest for the development of efficient negative ion sources and ion transport, in particular, for neutral beam heating in fusion research.

The electron detachment in H^--He collisions was studied in many theoretical and experimental works (e.g., see Ref. [1] and references therein), most of them being devoted to calculation and measurement of the total detachment cross sections. For the colliding system H^--He, experimental data on the total detachment cross sections in the ion energy range from 0.2 up to 40 keV have been reported [2–8]. In a few papers, experimental data on energy loss spectra [9] and cross sections for electron detachment with hydrogen atom excitation [10,11] have been reported. Experimental data on the energy spectra of detached electrons have been reported in Ref. [12] (relative values, low electron energies) and Ref. [13] (absolute values, broad electron energy range). Theoretical treatments of electron detachment are given in a number of papers [14–21]. These theories have led the authors to models of the detachment process described below.

The whole collision energy range can be divided into two regions. At low energies ($E_H^- < 2$ keV), the adiabatic approximation applies, and the detachment process can be considered to result from the promotion of the initial quasimolecular term to the continuum with formation of a quasistationary state statically or dynamically coupled with the continuum [14,15,18–20]. In the second region ($E_H^- > 5$ keV), the process can be considered as due to free-electron scattering on an atomic target [16,17,21]. A large difference between binding energies of the outer and core electrons (e.g., 0.75 and 13.6 eV for H^-) and, consequently, between their orbital dimensions, ensures that the zero-range potential approximation is an effective description of atomic fields [14]. Then the problem is reduced to a proper choice of the boundary condition imposed on the outer electron wave function at the atom position. Such approximations were successfully used in calculations of the total detachment cross sections in H^--He collisions at low ion energies [20], though a noticeable disagreement between the calculations and more recent experimental data [1] was found in the keV energy range.

The theoretical description of H^- is usually made in terms of *independent* loosely and tightly bound electrons, i.e., a split shell $1s1s'$, such that the processes with the inner tightly bound $1s$ electron do not affect the behavior of the outer loosely bound $1s'$ electron and vice versa. Only a few suppositions about the possible role of the electron correlations have appeared in the literature (e.g., Ref. [17]), but no detailed studies of their dynamical effect have been performed up to now. Probably this is for the following reasons. First, the independent electron approximation gives satisfactory agreement (within the experimental errors) with available experimental data on the total detachment cross sections. Second, up to now, it was not possible to perform exact quantitative calculations of ejected electron energy spectra, in which the correlation effects manifest themselves more distinctly than in the total cross sections. Such complex experimental and theoretical studies of electron detachment in H^--He collisions are reported in the present work.

II. EXPERIMENTAL TECHNIQUE

The doubly differential cross sections (DDCSs) for electron ejection have been measured using conventional electron spectroscopy [22,23]. Briefly, a H^- ion beam from a

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duoplasmatron entered a gas cell placed inside a cylindrical electrostatic mirror with an entrance angle $\theta_{\text{lab}}=54.5°$ and energy resolution $\Delta E/E=0.63\%$. Energy-analyzed electrons entered a detector consisting of a channeltron mounted behind a small conical deflector used to discriminate against spurious electrons. The incident beam current was measured with a Faraday cup and registered with an electrometer with output to an anlog-to-digital converter (ADC). The residual magnetic field was reduced to $H<10$ mG by μ-metal shielding and compensation of the field using three pairs of Helmholz coils. The absolute calibration of the measured doubly differential cross sections was made by determining the constants entered in the standard expression for the doubly differential cross section both from special control experiments and from normalization to the recommended data available in the literature. Special experiments using an electrostatic deflector before the collision chamber were performed to ensure that the fraction of electron signal produced by neutral atoms in a beam is negligible (less than 3%).

In this work, the measurements of the doubly differential cross sections for electron emission in H^--He collisions have been performed in the ejected electron energy range $E_e=2-40$ eV and in the incident ion energy range $E_{H^-}=1-10$ keV. The accuracy of the absolute values of the measured cross sections is estimated as ±15%. When comparison is possible, the data obtained show good agreement with the previous measurements of Risley [13].

III. THEORETICAL APPROACH

Our theoretical investigations employ the zero-range potential (ZRP) models for electron interactions with neutral H and He atoms in H^--He collisions and the Coulomb potential for electron interactions with H^+ in H^0-He collisions. The ZRP model has long been used to treat negative ion collisions [12,15,17,20]. The ZRP approximation is amenable to the molecular Sturmian theory [24], which is based on the three transformations: (i) the scale transformation of Solov'ev-Vinitsky [25], (ii) expansion of the total wave function in eigenfunctions of a Sturm-Liouville problem, and (iii) representation of the wave function in the form of the Fourier integral. In this approximation, the parameter $\omega=E(R)R^2$ is introduced instead of the conventional adiabatic energy $E(R)$, and the spatial motion is described by the dimensionless parameter $\boldsymbol{q}=\boldsymbol{r}/R(t)$. Then a Sturm-Liouville problem is furnished by the system of differential equations

$$[H_0(\boldsymbol{q})+\rho\nu(\omega)V(\boldsymbol{q})-\omega]S_\nu(\omega,\boldsymbol{q})=0 \tag{1}$$

with proper boundary conditions [26]. The values of internuclear distance at fixed potential energy $V(\boldsymbol{q})$ are taken as new eigenvalues $\rho_\nu(\omega)$, which are solutions of the equation

$$\varepsilon(\rho)\rho^2=\omega=\text{const}. \tag{2}$$

The corresponding Sturmian eigenfunctions $S_\nu(\omega,\boldsymbol{q})$ are defined for all values of ω, including negative, positive, and complex values. In contrast to the conventional adiabatic functions, the Sturmians do not depend on the internuclear distance. In the case of ZRP and Coulomb potentials the Sturmian method provides an exact solution of the time-dependent Schrödinger equation for collision systems with one active electron, because it includes effects of electron translation and rotation of the internuclear axis that have been omitted in most previous formulations. An important advantage of the Sturmian method is a very good convergence of solutions, such that only one or two Sturmian functions are usually needed to describe a particular collision process. The theory has an extraordinary energy range of validity, the lower limit of which is set by approximation of classical trajectories and the upper limit defined by neglecting penetration of the active electron into the He core. Thus, the Sturmian calculations can be considered as the best one-electron treatment of electron detachment at low and intermediate energies.

At low collision energies, the single-electron ionization process can be described by the advanced adiabatic approximation [27]. In this approximation, the energy $E(R)$ is replaced by the reciprocal function $R(E)$, so that the probability for electron ejection with energy E can be written as

$$P(E)=\frac{1}{2\pi v}\left|\frac{dR(E)}{dE}C^2(E)\exp\left(\frac{2i}{v}\int^E R(E')dE'\right)\right|. \tag{3}$$

Then the differential cross section in the center-of-mass system can be written as [4]

$$\frac{d\sigma}{dE}=\frac{4\pi|R(E)|^2\,\text{Im}\,R(E)}{\alpha(E)}K\left(\frac{\alpha(E)}{v}\right),$$

$$\alpha=2\int^E \text{Im}\,R(E')dE', \quad K(x)=e^{-x}(1-e^{-x}). \tag{4}$$

The advanced adiabatic approximation has been used in the present calculations of electron energy spectra in H^0-He collisions. Moreover, using Eqs. (3) and (4) one can determine parameters of the promoted diabatic quasimolecular term, such as $\text{Re}\,R(E)$ and $\text{Im}\,R(E)$, directly from experimental data on electron energy spectra [22]. The relations connecting the center-of-mass and laboratory systems are given in Ref. [28]. To compare experimental data and calculations, a H^- ion energy of 2 keV has been chosen, which falls into the "adiabatic region" [18].

IV. EXPERIMENTAL RESULTS

Figure 1 shows the doubly differential cross sections for electron ejection in H^--He collisions as a function of electron energy measured in the ion energy range $E_{H^-}=1-10$ keV. Four main components can be discerned in the experimental electron energy spectra, namely, a continuous part of the spectrum, a cusp at $\boldsymbol{v}_e=\boldsymbol{v}_{H^-}$, and two structures associated with decay of autoionizing states $2s^2$, $2s2p$, $2p^2$, etc. of H^- ions and He atoms. The data obtained at 1, 2, 5, and 10 keV agree well with the earlier data [13] for the nearest angle $\theta=60°$, which serves as a check on our absolute cross sections.

Figure 2 shows the total cross sections for excitation of the autoionization states of H^- and He as a function of ion

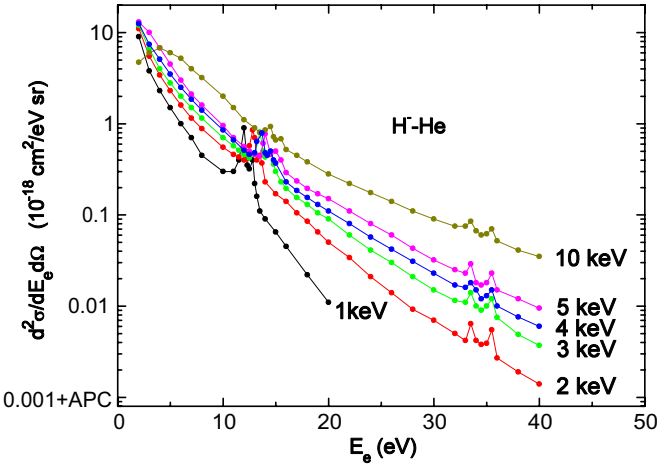

FIG. 1. (Color online) Energy spectra of electrons ejected in H^--He collisions at $\theta_{lab}=54.5°$. The structures near 12 eV are autoionizing states of H^- and the structures near 12 eV are autoionizing states of He.

energy determined from the peak areas in assumption of isotropic angular distribution of ejected electrons. Again the data for H^- agree with those of Ref. [13] within the experimental accuracy. We know of no other data for comparison of the data for autoionization states of He excited by H^- impact.

V. DISCUSSION

Now we proceed to the discussion of the continuous part of the spectrum, which carries detailed information on the most important mechanisms of electron detachment

Figure 3 shows the measured and calculated electron energy distributions for 2 keV H^- ion impact on He. One can see that the model Sturmian calculations for H^- agree well with the measurements only at low electron energies ($E_e<8$ eV), while at higher energies they differ strongly

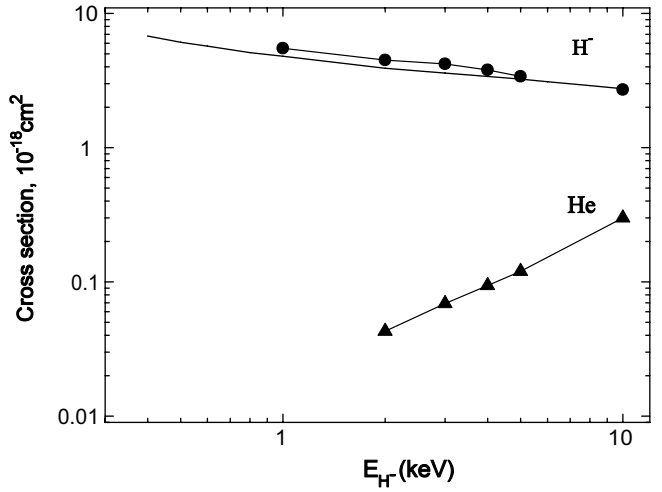

FIG. 2. Cross sections for excitation of autoionization states of H^- and He. Solid line (Ref. [13]); full circles and triangles, this work.

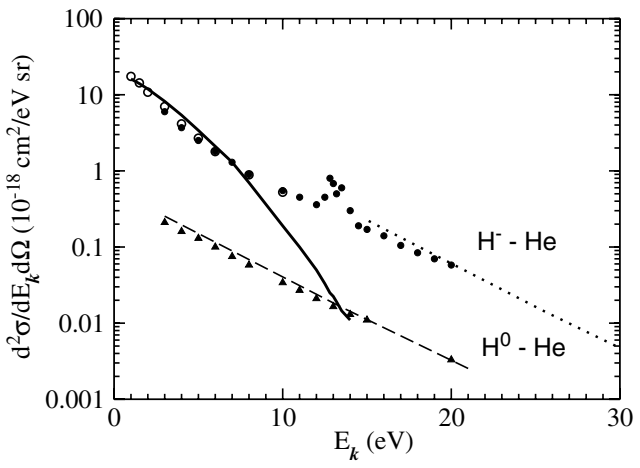

FIG. 3. Energy spectra of electrons ejected in H^--He and H^0-He collisions. Experimental data: filled circles, this work H^--He; open circles, Risley (Ref. [13]); triangles, this work H^0-He. Calculations: solid line, H^--He; dashed line, H^0-He. The dotted line shows the calculated values for H^0-He shifted to the right by 12.85 eV.

from the experimental data both in the absolute values and in the shape of the energy distribution. For the H^- spectra the theoretical cross sections drop much faster than exponential above 10 eV whereas the measured DDCSs decrease exponentially with a constant slope on a semilogarithmic plot. The comparison of the data given in Fig. 3 supports the idea that the conventional mechanism of the loosely bound $1s'$ electron "squeezing out" is mainly responsible for ejection of electrons with energies below 8 eV, while some other mechanism, in which the tightly bound $1s$ electron is involved, plays a dominant role above 10 eV. The simplest independent particle picture supposes that the DDCSs in the high-energy region are due to promotion of $1s$ electron and unaffected by the spectator $1s'$ electron.

To check this supposition, we compared the data obtained for H^- with calculations and experimental data [22] for neutral atom H impact on He. In the H^0-He (as well as in the H^--He) system the Coulomb barrier is absent [29], so that the saddle point mechanism for the inner electron excitation does not work and the only possible mechanism is superpromotion (S ionization) [30]. The classical interpretation of S ionization is based on the topology of the electron motion. At close approach of colliding systems, a united atom centrifugal barrier appears. This barrier keeps the electron out of the region between the nuclei, so that the trajectory along the line between the nuclei becomes unstable. Oscillation of the electron along this unstable trajectory transfers energy from nuclear to electron motion up to the values sufficient for electron emission.

The contribution of S ionization can be estimated using the advanced adiabatic approximation [31] Eq. (4). As seen from Fig. 3, the calculations and measurements for H^0-He agree where experimental data are available. The agreement in the slopes of curves for H^- and H^0 supports the idea that S promotion of a $1s$ electron is responsible for the appearance of fast electrons seen in the cross sections for H^--He. However, the DDCSs for H^0-He are about an order of magnitude

smaller than those for H^--He. Thus, this is really the $1s'$ electron that is emitted, though its emission process is governed by behavior of the inner $1s$ electron. In other words, a strong dynamical correlation of the $1s$ and $1s'$ electrons is responsible for formation of the high-energy part of the electron energy spectrum. It should be noted that quite a good agreement between the data for H^- and H^0 is obtained when shifting the H^0-He curve to the right by an amount equal to 12.85 eV, the difference between the ionization energies of H^- and H^0. So the transition frequency $\omega=\Delta E(R)/h$ rather than the electron energy is the main characteristic of the correlation process.

This process can be described in terms of promotion the two-electron state $1s1s'$ to the continuum via a diabatic term. The parameters characterizing this term have been determined in this work both from *ab initio* calculations using the ZRP model [32] and from the experimental data using Eq. (4). Comparison of the results obtained for $E(\mathrm{Re}\,R)$ is given in Fig. 4, showing quite good agreement between theory and experiment.

Now the question arises about the mechanism responsible for large energy transfer to the loosely bound $1s$ electron of H^-. Some analogy can be traced with the situation occurring in ionization of weakly bound states (Rydberg atoms, negative ions) in a microwave field [33–36]. In our case, a similar field is created by oscillations of the inner $1s$ electron in the course of S promotion.

The revealed mechanism associated with dynamical correlated motion of two electrons can be called correlated electron detachment. Preliminary estimates based on integration of the corresponding parts of energy spectrum show that the correlated electron detachment yields about 30% of the differential detachment cross section at 2 keV, 90% of which results in excitation with ionization and 10% in double ionization. Such sharing is consistent with the data obtained in scattering experiments [9]. More detailed quantitative description of the correlated electron detachment requires solution of the very difficult two-electron problem.

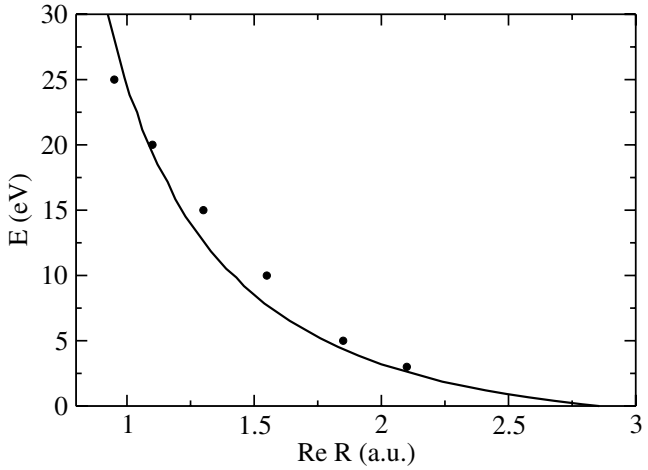

FIG. 4. The function $E(\mathrm{Re}\,R)$ for the HHe^- superpromotion state. Solid curve, model calculation; full circles, the values extracted from experimental data.

The present observations suggest that further theoretical and experimental investigations for negative ions would prove fruitful. In fact, our interpretation suggests that the dynamical correlation effect would be present for almost all negative ion species where superpromotion provides the main avenue for electron detachment in both the negative ion and the parent neutral atom.

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