Orbiting charge-transfer cross sections between He^+ ions and cesium atoms at near-thermal ion-atom energies

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The energy dependence of the total charge-transfer cross section for the collision of helium ions with cesium atoms at epithermal energies (0.1 to 1 eV) has been measured. It was observed that the cross section increases with $E^{-1/2}$. This can be attributed to orbiting collisions. The charge-transfer cross section is expressed as the product of the classical orbiting cross section and a quantum-mechanical charge-transfer probability which is shown to be energy independent.

The effect of orbiting on the total cross section for atom-atom collisions has been studied in a large number of experimental[1,2] and theoretical investigations.[3,4] In comparison, less attention has been paid to the influence of orbiting in ion-atom collisions. One special feature, when one of the colliding systems is an ion, is the long-range polarization force, which introduces an attractive interaction energy that is proportional to the polarizability of the atom.

In this paper we report on the application of an ion storage method[5] to the study of charge-transfer reactions between highly polarizable cesium atoms and rigid helium ions at low relative collision energies. Total charge-transfer cross sections are presented for the He^+-Cs system in the energy range of 0.1 to 1 eV. In this low-energy range the collisional interaction time is long against the orbital period of the active electron, and coupled molecular state approximations can be used to describe the collision if the polarization of the atom in the field of the ion is taken into account. However, so far no detailed calculations have been carried out with this model. Alternatively, the classical concept of orbiting collisions according to Langevin[6] can be employed and used to explain the energy dependence of the measured collision cross sections.

The experiment investigates the near resonant charge-transfer process between stored He^+ ions and cesium atoms. The reaction proceeds along the following near resonant channels when using the separated atom notation to describe the collision:

$$\mathrm{Cs}(6s\,^2S_{1/2}) + \mathrm{He}^+(1s\,^2S_{1/2}) \rightarrow \begin{cases} \mathrm{He}^*(1s2p\,^1P_1) + \mathrm{Cs}^+(^1S_0) - 0.52\ \mathrm{eV} \\ \mathrm{He}^*(1s2p\,^3P_{2,1,0}) + \mathrm{Cs}^+(^1S_0) - 0.27\ \mathrm{eV} \\ \mathrm{He}^*(1s2s\,^1S_0) + \mathrm{Cs}^+(^1S_0) + 0.08\ \mathrm{eV} \\ \mathrm{He}^*(1s2s\,^3S_1) + \mathrm{Cs}^+(^1S_0) + 0.88\ \mathrm{eV}\ . \end{cases} \tag{1}$$

A large number of channels of a different nature is also available at the same time:

$$\mathrm{Cs}(6s\,^2S_{1/2}) + \mathrm{He}^+(1s\,^2S_{1/2}) \rightarrow \mathrm{He}(1s^2\,^1S_0) + \mathrm{Cs}^{+*}[5p^5(^2P)nl] + \Delta E_{n,l}\ , \tag{2}$$

where $n = 6-10$, and $l = s$, p, and d. In this reaction an electron is transferred from the closed $5p^6$ shell of cesium to the $1s$ shell of helium. When orbiting occurs, the collision partners spend a relatively long time close to each other so that the probability of charge transfer is high.

The experimental arrangement has been described previously.[7] Briefly, it consists of an ion trap located in a glass vacuum chamber with a base pressure of about 1×10^{-10} Torr and a cesium atomic beam produced by a reflux-type oven. The cesium atomic beam enters the trap through windows cut into the ring electrode. Helium is introduced into the system by heating a quartz leak. He^+ ions are generated by pulsed electron bombardment and stored in the rf quadrupole trap for many seconds. The stored He^+ ions interact with the cesium beam and the ion loss rate yields the total charge-transfer cross section.

The rf ion trap was operated in the symmetric frequency mode. The well depth of the symmetrically operated ion trap was changed by changing the operating voltages. In this way the charge-transfer cross section was studied over the energy range of 0.1 to 1 eV. In this kind of operation the ion oscillation frequency changes for each well depth and a broadband detection scheme has to be used. An ion signal is obtained by coherently exciting the ions at the sideband frequency $\Omega+\omega$ and detecting the voltage induced by the cooperative ion motion in a large resistor. After amplification, heterodyning, and squaring of the signal, it is recorded by a fast analog-to-digital converter and an on-line computer. The ion signal was calibrated in terms of absolute ion numbers by measuring the space-charge shift of the signal.[8] The number of stored He^+ ions left in the trap after a fixed interaction time with the cesium beam yields the total charge-transfer cross section of interest. The appearance of Cs^+ could not be measured as a consistency check since the vastly different masses of He^+ and Cs^+ did not allow simultaneous trapping of both species.

The relative collision energy is given by the average energy E of the stored He^+ ions, since a thermal atomic cesium beam was used. The average ion velocity $\bar{v}$ is evaluated from the relation[9]

$$\bar{E} = 0.10eD = m\bar{v}^2/2 \ , \tag{3}$$

where D is the depth of the potential well of the ion trap and e is the electron charge. Care was taken to only partially fill the trap with He^+ ions. This reduces the deformation of the potential well by space-charge effects and ion losses due to ion-ion collisions. For such an unsaturated trap the ion-atom charge transfer is the dominant process and is described by the ion loss rate $1/\tau$ expressed as

$$1/\tau = \tilde{N}_{He}\bar{v}(\sigma_{He} - \sigma_A) + \tilde{N}_{Cs}\sigma_{Cs}\bar{v} \ . \tag{4}$$

The two terms in Eq. (4) illustrate the two competing charge-transfer processes. The first term is due to the resonant charge-transfer process in the He^+-He system, and the charge-transfer cross section σ_{He} reflects the interaction of the stored He^+ ion cloud with the helium background gas. The second term describes the near resonant charge-transfer process characterized by the charge-transfer cross section σ_{Cs} in the He^+-Cs system and occurs in the presence of the cesium atomic beam. The cross section σ_A is a measure of the acceptance A of newly created helium ions in the trap by charge transfer of neutral helium atoms with previously stored ions. We define A as

$$A = \sigma_A/\sigma_{He} \ , \tag{5}$$

which is essentially the ratio of the number of ions stored to the number of ions formed.

The experiment was carried out in two steps. First, the acceptance A of the trap was measured using the known charge-transfer reaction between helium atoms and helium ions for calibration. For this measurement the cesium beam is turned off and only the first term in Eq. (4) contributes. The mean lifetime τ of the stored He^+ ions is determined as a function of both the relative velocity $\bar{v}$ and the helium pressure. The energy dependence of A is obtained using the results of Mahadevan and Magnuson[10] for the symmetric charge-transfer cross section σ_{He}. Figure 1 displays the values for σ_A and A obtained for a partially filled ion trap operated in the symmetric frequency storage mode. It illustrates increasing acceptance with increasing well depth or average ion energy E.

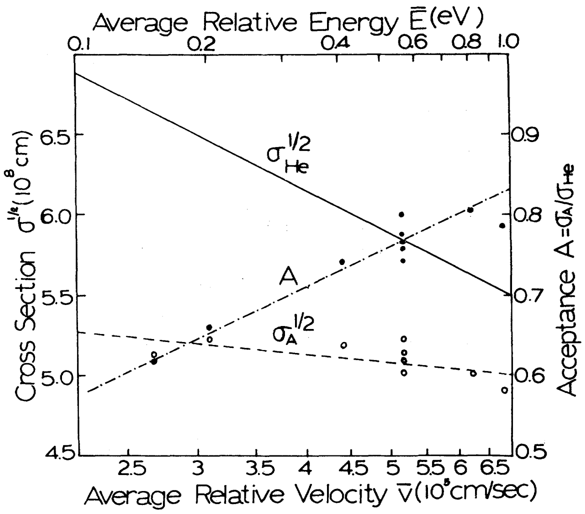

FIG. 1. Charge-transfer cross section σ_{He}, acceptance cross section σ_A, and ion trap acceptance factor A as a function of the average ion energy.

In the second step of the experiment the cesium atomic beam was turned on. Now both terms in Eq. (4) contribute. The cesium density in the trapping region was obtained from the temperature of the cesium oven using also a hot wire detector for reproducible adjustment. The value of the cesium density in the trapping region was $N_{Cs} = 2.7 \times 10^7$ atoms/cm^3.

From the second term the He^+-Cs charge-transfer cross section is obtained. Again the measurements were made for several well depths or average ion-atom velocities. The results are shown in Fig. 2. The total He^+-Cs transfer cross section σ_{Cs} increases with decreasing ion-atom energy. The cross-section value is inversely proportional to the square root of the average ion energy. This behavior demonstrates the occurrence of orbiting charge-transfer collisions in this energy range.

In the energy region below 1.0 eV the attractive charge-induced dipole forces between the ion and the atom become important. The orbits of the slow particles are no longer rectilinear with constant velocity as for higher-energy collisions. Under certain conditions one of the collision partners even revolves about the other particle many times before it moves away. When no strong chemical interaction exists between an ion and a neutral atom, the interaction potential between these particles can be assumed to be of the form[3,11]

$$V(R) = \begin{cases} -\dfrac{\alpha e^2}{2R^4}, & R > a \\ \infty, & R < a \ , \end{cases} \tag{6}$$

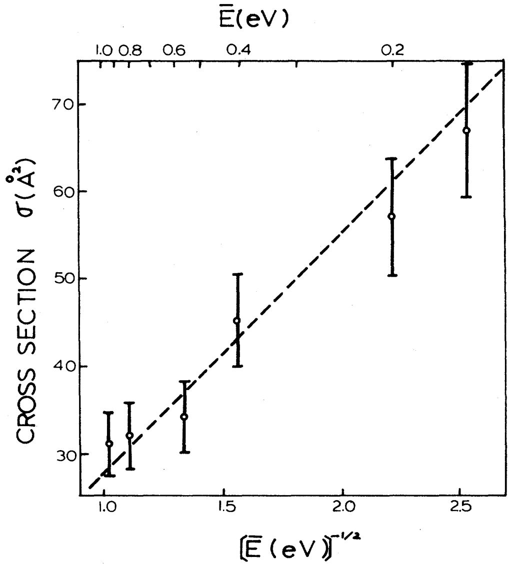

FIG. 2. Plot of the measured helium ion-cesium atom charge-transfer cross section $\sigma_{He^+\text{-}Cs}$. The uncertainty in the energy is about 10% and is now shown in the error bars. The dashed line is the best fit to the experimental points.

where R is the particle separation, a is the equilibrium separation, and α is the polarizability of the atom. For He^+-Cs system, α is given as (56 ± 8) Å^3. This value is the average of the three most reliable measurements[12–14] of α, which are tabulated for the cesium atom.[15] When the centrifugal force is included, the effective potential for radial motion is

$$V_{\text{eff}}(R) = V(R) + L^2/2mR^2 \tag{7}$$

for a relative angular momentum L and reduced mass of the collision partners m. If the impact parameters b and the relative kinetic energy E are used instead, Eq. (7) becomes

$$V_{\text{eff}}(R) = V(R) + b^2E/R^2 \; . \tag{8}$$

The particle orbit depends on the value of the impact parameter and the relative kinetic energy. For high values of b and E, V_{eff} is repulsive. But for low values of b and E, a local maximum V_{eff} may exist. For orbiting to occur, the radial velocity, which is proportional to $[E - V_{\text{eff}}(R)]^{1/2}$, must vanish at a particle separation corresponding to an accessible maximum of $V_{\text{eff}}(R)$. For the induced dipole interaction potential $V(R)$, the critical impact parameter is given by

$$b_c = \left[\frac{2\alpha e^2}{E}\right]^{1/4} \; . \tag{9}$$

The orbiting cross section is defined as

$$\sigma_{\text{or}} = \pi b_c^2 = \pi \left[\frac{2\alpha e^2}{E}\right]^{1/2} \; . \tag{10}$$

TABLE I. Comparison of charge-transfer cross sections as obtained from orbiting calculations and from the analysis of the experimental results.

$\bar{E}$ (eV)	b_c (Å)	σ_{or} (Å^2)	σ_{expt} (Å^2)	$\eta = \dfrac{\sigma_{\text{expt}}}{\sigma_{\text{or}}}$
0.155	10	314	67 ± 8	0.21 ± 0.03
0.202	9.5	283	58 ± 6.8	0.20 ± 0.03
0.406	7.9	196	45 ± 5.3	0.23 ± 0.03
0.560	7.3	167	34 ± 4.1	0.20 ± 0.03
0.808	6.7	141	32 ± 3.8	0.23 ± 0.03
0.950	6.4	129	31 ± 3.7	0.24 ± 0.03

For the He^+-Cs system the critical impact parameter is about 6.3 Å for a relative kinetic energy of 1 eV and the orbiting cross section at low relative kinetic energies is

$$\sigma_{\text{or}} = (126 \times 10^{-16}/E^{1/2}) \; , \tag{11}$$

with σ in units of cm^2 and where E is in units of eV. Whenever orbiting occurs, the colliding particles spend a relatively long time close to each other so that there is a good chance for any reaction between these particles to occur. Let η be the probability of the charge-transfer reaction to take place while orbiting occurs. Then the charge-transfer cross section is given by

$$\sigma_{\text{or}} = \eta\sigma_{\text{or}} = \eta\pi \left[\frac{2\alpha e^2}{E}\right]^{1/2} \; . \tag{12}$$

This energy dependence of the charge-transfer cross section at low energies is verified by the experimen-

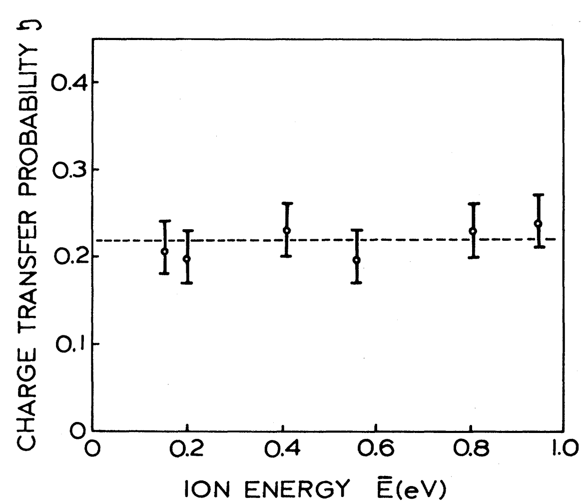

FIG. 3. Plot of the charge-transfer probability during orbiting for different ion energies.

tally measured values. The ratio of the experimental charge-transfer cross section and the orbiting cross section determines the charge-transfer probability during orbiting. The values of σ_{Cs}, σ_{or}, and η are given in Table I. Figure 3 shows that the values of η are about 0.22 and that they are energy independent at the low-energy region considered.

We have demonstrated that an orbiting type of charge-transfer collision occurs for the Cs-He^+ system below ion-atom energies of about 1 eV. The ions follow a curved path and spiral inwards toward the atom until the repulsive potential core reverses the trend. The product of the classical orbiting cross section and the quantum-mechanical charge-transfer transition probability η yields the charge-transfer cross section for collisions in this low-energy region. The charge-transfer probability during orbiting is observed to be energy independent.

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