14 A simple argument which illustrates this is as follows: The rate of electron transfer [process (4) in text] is given by

$$\frac{d[B^{\bullet}]}{dt} = k [A^*][B],$$

where k is the rate constant and $[A^*]$ and [B] are the concentrations of A^* and B, respectively. However if the ion core of the A^* atom plays no part in the interaction we may write this as

$$\frac{d[B^{-}]}{dt} = k[e][B],$$

since $[A^*] = [e]$. But this is just the rate of electron

attachment to molecule B [process (5) in text].

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Single and Double Charge Transfer in C⁴⁺-He Collisions*

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Cross sections for single and double charge transfer from C^{4+} incident on He have been measured and calculated for relative velocities between (0.2 and 1.2) ×10⁸ cm/sec. The double-charge-transfer cross section rises with decreasing energy and is predicted to be nearly two orders of magnitude larger than that for single charge transfer at the lowest velocity. The single- and double-charge-transfer cross sections are experimentally found to be equal at a velocity of 6.5×10^7 cm/sec with a value of 2.7×10^{-16} cm².

Multiply charged ions of common atoms are of considerable interest in high-temperature and nonequilibrium plasmas. Because of large cross sections, charge exchange between these ions and other atomic constituents in the plasmas plays a significant role in important plasma properties such as charge-state distribution, resistivity, and energy transfer. In the interstellar medium, charge exchange is important in determining which ion states are expected.¹ In tokamak plasmas charge exchange of multiply charged ions is important for the properties mentioned above; in addition, for injected neutral beams, charge exchange with multicharged impurities may be instrumental in stopping the injected beam at the outer edge of the plasma, thus destroying the needed plasma heating.² An additional area where such charge-transfer cross sections are

important is in the search for x-ray lasers, where charge exchange into an excited state of a multicharged ion could provide a mechanism for pumping the required excited-state population.³

Because of these several important areas of application, it is necessary to develop better quantitative knowledge of the collision processes involving multiply charged ions. In the case of charge exchange at relatively slow velocities, little experimental work exists and dependable theoretical predictions are also scarce; but both experiments⁴ and theory are actively being pursued. In the course of recent investigations, some of the qualitative ideas held in general use for estimating effects of charge exchange with multicharged ions have proven to be false in many cases.

Usually, it is assumed that for slow collisions

of multicharged ions with neutral atoms, $V_{rel} < 2 \times 10^8$ cm/sec, single charge transfer is the dominant process (see Presnyakov and Ulantsev⁵ for example). In resonant charge exchange this is not always true (see Bayfield and Khayrallah⁶ for the example of He⁺⁺ on He). The purpose of this Letter is to point out an interesting nonresonant case in which the cross section for double charge transfer, C⁴⁺ +He⁺ C²⁺ +He²⁺, is considerably larger than that for single charge transfer, C⁴⁺ +He⁺C³⁺ +He⁺.

In general for collisions in which the nuclear velocity is slower than the electron orbital velocity a quasimolecule is formed during the collision, and the collision can best be understood in terms of the molecular potentials of the reactant and product states. For the case at hand, HeC^{+4} , we have used configuration-interaction wave functions to calculate the potential energy curves and the radial $(\partial/\partial R)$ and rotational (L_v) coupling between the various molecular states. The potential energy curves so derived are shown in Fig. 1. The principal features of the curves are the avoided crossing between the ${}^{1}\Sigma_{4}$ (initial) and ${}^{1}\Sigma_{3}$ (double charge transfer) states at $R \sim 3.5 a_0$, the close approach of the ${}^{1}\Pi$ and ${}^{1}\Sigma_{3}$ states at small R values, and the avoided crossing between the $^{1}\Sigma_{1}$ and $^{1}\Sigma_{2}$ states at $R \sim 2.7a_{0}$.

From the potential curves shown in Fig. 1, it can be seen that for a low-energy inelastic collision, the approaching particles will only have sufficient energy to transit the first avoided curve crossing to the ${}^{1}\Sigma_{3}$ molecular state and produce double-charge-transfer products. As the collision energy is increased, however, the particles will be increasingly able to transit further the second avoided crossing to the ${}^{1}\Sigma_{2}$ state and pro-



FIG. 1. Singlet potential-energy curves for the various electronic configurations as a function of internuclear separation.

duce single-charge-transfer products. Also, at the higher energies rotational coupling plays a role and single charge transfer proceeds via Σ - Π interactions. At the collision energies investigated, all the single-charge-transfer products were found to be produced in the $C(1s^22p)^{+3}$ state, the avoided crossing between ${}^{1}\Sigma_{2}$ and ${}^{1}\Sigma_{1}$ being too large to allow flux into the $C(1s^22s)^{+3}$ channel. The next higher single-charge-transfer channel would be $C(1s^23s)^{3+}$. However, the avoided crossing with the initial channel is at $R \sim 35 a_0$ where the coupling will be negligible. Hence, from the potential curves shown in Fig. 1, we would expect that the double-charge-transfer products will dominate at the lower collision energies, while at the higher energies singlecharge-transfer products will.

The cross sections for the various product states were calculated by solving the coupled equations assuming classical motion of the particles with straight-line trajectories. We should note that the calculated cross section for the primary process (double charge transfer) should be accurate to approximately $\pm 25\%$, while the cross section for the secondary process (single charge transfer) which proceeds via the doublecharge-exchange intermediary state will be less accurately defined—probably only accurate to approximately $\pm 50\%$.

An experiment measuring these cross sections has also been performed with the apparatus and method discussed by Crandall.⁷ Ions are formed in a dc arc of a cold-cathode Penning-type ion source capable of producing high-charge-state ions and selecting ions of given mass and charge.⁸ The extracted C^{4+} ions are transported to a collision chamber where a highly collimated beam is passed through a short cell and into a parallelplate electrostatic analyzer which deflects particular charge states onto the desired channel electron multiplier. By measuring the distribution of ions in final charge states with and without helium in the collision cell, the single- and double-charge-transfer cross sections were obtained. Experiments were performed at those energies for which ions could be extracted from the source (5 to 20 kV extraction potential).

The results are shown in Fig. 2 together with the calculated values and the single-chargetransfer data from a previous experiment by Zwally and Koopman.⁹ For single charge transfer, quantitative agreement among the three cases is quite poor. The measurements of Zwally and Koopman are about a factor of 2 greater than



FIG. 2. Cross sections for single and double electron capture from C⁴⁺ incident on helium. Solid curve, present calculation of σ_{42} for C⁴⁺ + He \rightarrow C²⁺ + He²⁺. Crosses, present experiment for σ_{42} . Dashed curve, present calculation of σ_{43} for C⁴⁺ + He \rightarrow C³⁺ + He⁺. Open circles, present experiment for σ_{43} . Closed circles, σ_{43} from Ref. 9.

the present measurements, which are in turn about a factor of 2 greater than the present theory. The work of Zwally and Koopman was a carefully performed experiment which relied on a pulsed ion source resulting in some severe technical difficulties not encountered in the present experiments. Except for the ion source, the two experiments are so similar in technique that discrepancies as large as shown in Fig. 2 are difficult to explain. In both experiments the metastable content (excited states of the He-like C^{4+} ion) is unknown but expected to be small on the basis that metastable formation should be similar to C^{5+} formation which is known to be very small in both sources. Nevertheless, the discrepancy between experiments could be due to different metastable content; experiments are planned to investigate the metastable content of beams from the present ion source. Zwally and Cable¹⁰ have also reported measurements for B^{3+} +He which were obtained with a different dc ion source. A check of these results with the present apparatus gave agreement within 20%.

For single charge transfer, the present data are in substantially better agreement with calculated values than the previous measurements. Nevertheless, this discrepancy between present experiment and theory is a little larger than the sum of estimated uncertainties. For the experiment, the anticipated uncertainty of about $\pm 25\%$ is roughly equal to the separation of the two data points at V_{rel} of 8×10^7 cm/sec shown in Fig. 2. These two data points were separated by several months in time with complete disassembly of the collision chamber and a change of the helium-gas source.

For double charge transfer, the agreement between calculated and measured values is satisfying. As mentioned above, the double-chargetransfer process is more accurately calculated so that improved agreement over the singlecharge-transfer case is anticipated. The theory predicts extreme dominance of double electron capture at low velocities, and the available data support this conclusion. Clearly, any assumption that single-electron-capture cross sections are always larger than double-electron-capture cross sections is unwarranted. A reasonably detailed consideration of the molecular nature of the colliding system is required before even qualitative estimates of such processes are meaningful.

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