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Electron transfer between He-like ions and He

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Single- and double-electron-transfer cross sections have been measured for B^{3+}, C^{4+}, N^{5+}, and O^{6+} in collision with He at velocities between 0.5 and 1.2×10^8 cm/sec. The single-electron-capture cross sections peak near 15×10^{-16} cm^2 for each case except C^{4+}, where single capture is anomalously low. The double-capture cross sections are about 3×10^{-16} cm^2, except for B^{3+}, where the highest observed value is 1.5×10^{-16} cm^2. These measurements for B^{3+} and C^{4+} compare well with existing experiments and theory, except for C^{4+} single capture. Within the narrow range tested, C^{4+} and B^{3+} cross sections exhibit variation with velocity, but N^{5+} and O^{6+} cross sections remain constant.

INTRODUCTION

Electron capture by multicharged ions in collision with neutral atoms is a fundamental process of intrinsic interest. In addition, the significance of such electron transfer to plasma behavior and in diagnostics is gaining recognition.[1] Because of its comparatively large cross sections, electron transfer is sometimes the fastest recombination process affecting ion charge state of the multicharged ions created in high-temperature plasmas — particularly in some astrophysical situations where the high ion charge states are produced by photon impact.[2,3] For multicharged ions, electron transfer occurs primarily into excited states which will radiate, dissipating energy and presenting a useful source of light for plasma diagnostics. Additionally, electron capture into excited states is potentially useful to the development of short-wavelength coherent light sources (x-ray lasers).[4-6] Helium is an interesting collision target since it is one of the neutral species frequently encountered in environments where multicharged ions exist. Also, it is readily formed into a collision target and has an electronic structure simple enough to treat in collision models.

Calculations of electron transfer at velocities below 2×10^8 cm/sec are usually based on a quasimolecular model in which the colliding system is represented in terms of molecular stationary states. For multicharged ions, the initial state of the ion plus neutral atom is slightly attractive at large internuclear separation, while the states resulting from electron transfer consist of two ions which have long-range Coulomb repulsion. As the internuclear separation decreases, the potential of the two-ion states increases, resulting in avoided crossings of these states with the initial state of ion plus neutral. For ions of higher charge, there are usually more avoided crossings. Electron transfer is a transit of the colliding system from the initial state to one of the two-ion final states, and is most likely at the avoided potential curve crossings. In the Landau-Zener model, the transfer occurs only at these crossings, but more detailed calculations that include couplings away from crossings are generally necessary for accurate results.

This approach to calculations has been applied for cross sections of B^{3+}, C^{4+} + He (Refs. 7, 8); and some experimental data[8-10] already exist for these systems. The present work verifies previous results and extends experimental studies to higher charge states and to the capture of two electrons.

EXPERIMENTAL METHOD

A schematic diagram of the apparatus is presented in Fig. 1. This apparatus has been described in detail previously.[11,12] A beam of ions was selected at the adjustable slit for mass/charge, was passed through a gas cell, and was electrostatically charge-analyzed after the gas cell. Cross sections were obtained from the expression

$$N_f = N_q \sigma_{qf} n l ,$$

where N_f is the increase in the number of ions in a given final charge state f, due to addition of He to the gas cell; N_q is the number of incident ions in charge state q; n is the He density in the gas cell; and l is the gas cell effective length. Precise determination of all these quantities was discussed in

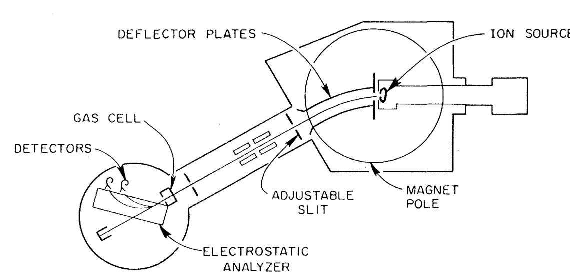

FIG. 1. Schematic of the apparatus.

Ref. 11.

Figure 2 shows the count rate at one of the detectors for 80-keV N^{5+} incident on 1.04×10^{-3} Torr of He in the cell, and with the cell evacuated (less than 3×10^{-6} Torr). Cross sections can be extracted directly from Fig. 2, but were obtained by two other procedures. For double electron transfer, cross sections were determined by using one detector and changing the analyzer voltage so as to detect alternately the primary beam of charge q, and the two-electron capture charge component of charge $(q-2)$. Most of the single-electron-transfer data were acquired using both detectors, so that the the ions of the initial charge state q were incident on one detector and, simultaneously, the ions of reduced charge $(q-1)$ were incident on the other (see Ref. 11).

The uncertainties pertinent to the experiment are the same as those discussed in Ref. 11. The reproducibility of the present data is not as good as that of previously reported data.[11] For the present single-electron transfer, five repeats of a particular data point gave a standard deviation of ±6.4%. A reproducibility of ±13% (approximately 90% confidence level) is taken to apply to all single-electron capture data. Table I lists a summary of the uncertainties. Systematic uncertainties have been estimated conservatively so that the absolute error should be within the limits to a high confidence — equivalent to 90% confidence level on statistics.

TABLE I. Summary of uncertainties.

Source	Single transfer (%)	Double transfer (%)
Reproducibility (90% CL)	±13	±15
Pressure + conductance	±10	±10
Temperature (±8 °C)	± 3	± 3
Effective length of gas cell	± 5	± 5
Gas purity (maximum effect on cross section)	± 5	± 5
Incident beam (N_q)	± 4	± 7
Relative counting efficiency	± 5	± 5
Quadrature sum	±19%	±22%

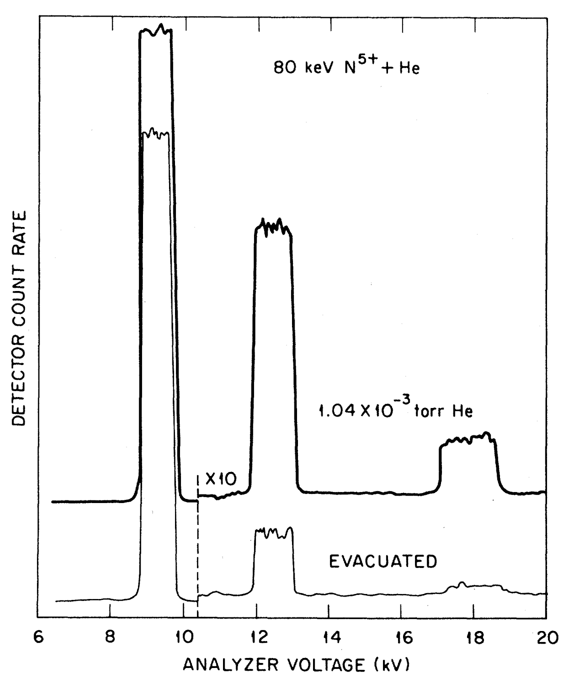

FIG. 2. Count rate at one detector as a function of analyzer voltage for incident N^{5+}; fine line is background, heavy line is with 1.04×10^{-3}-Torr He in gas cell.

RESULTS AND DISCUSSION

A. C^{4+} + He

Figure 3 presents the available data and theory for single and double electron capture for C^{4+} + He. With the exception of the data of Goldhar *et al.*,[13] these data have been discussed previously.[8] The calculations[7,8] employ accurate *ab initio* molecular potentials, and evaluate both radial and rotational

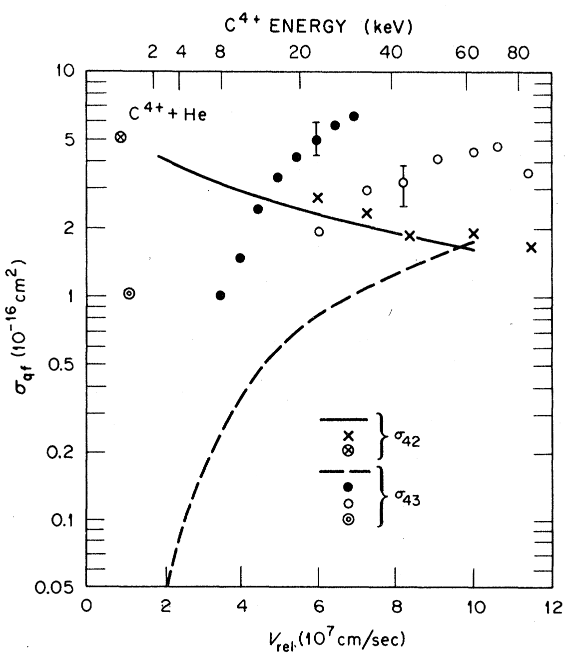

FIG. 3. Single- and double-electron capture by C^{4+} incident on He. Open circles, present data for single transfer σ_{43}; crosses, present data for double transfer σ_{42}; solid circles, σ_{43} from Ref. 9; circled point and circled cross, σ_{43} and σ_{42}, respectively, from Ref. 13; solid curve and dashed curve, calculation of σ_{43} and σ_{42}, respectively, from Refs. 7 and 8.

coupling for collision distance, $-10a_0 < r < 10a_0$ for the states considered. At the lowest velocities, σ_{42} is larger than σ_{43} due to a strong avoided crossing at internuclear separation $r \simeq 2.7a_0$, where the $^1\Sigma_4$ state tending to $C^{4+} + He$ interacts with the $^1\Sigma_3$ state tending to $C(1s^2 2s^2)^{2+} + He^{2+}$. The double capture is dominated by this one potential curve crossing, and the theory and experiments are in excellent agreement even though double-capture excited states having crossings about $r \simeq 5a_0$ have been neglected. For double capture, the initial to final-state coupling is strong only for small r so that the neglected crossings near $r = 5a_0$ are expected to be diabatic for velocities above 2×10^7 cm/sec. For σ_{43}, a clear discrepancy exists between present data and the theory. The calculation involves several states with both radial and rotational coupling. Most single transfer must proceed through transfer first to the $^1\Sigma_3$ double-capture potential curve, then by a subsequent transfer to a single-capture molecular curve. Thus, the σ_{43} cross section is small and difficult to calculate. The theory omitted electron transfer to the $C(1s^2 3s)^{3+} + He^+$ final state (and higher C^{3+} states) because the molecular curves cross at $34a_0$, and the coupling is so weak that the colliding system should transit diabatically through the crossing with no probability of transfer. This assumption may not be fully valid at the lowest collision velocities, since even small coupling of the states at a large radius could contribute significantly to the cross section. However, for the velocities of the present experiment, any inadequacy of the theory for single capture is probably due to incomplete description of the coupling of states which were included.

The independent measurements of σ_{43} do not agree. The work of Zwally and Koopman[9] relied on a pulsed discharge ion source which produced up to 10^5 C^{4+} particles in 2 μsec at the final detector. Performing an experiment with such an ion beam is difficult. The most easily postulated reason for discrepancy between our work and that of Zwally and Koopman is that the number of incident particles in their experiment was underestimated, due either to nonlinearity of the detector or failure to transmit all of the incident ions to the detector surface. Their experiment was carefully analyzed and performed, and they report no evidence for nonlinearity of detection for the intense short-duration pulses. However, all aspects of their work and our experiments are highly similar except for the type of detector and nature of the ion source. The B^{3+} work of Zwally and Cable[10] used the same apparatus as the C^{4+} work of Zwally and Koopman, except that the ion source was replaced by an electron impact type source which produced low-current continuous beams of B^{3+}. For the B^{3+} case, disagreement with present results will be shown to be less than the experimental uncertainties.

As a further test of the discrepancy between present results and those of Zwally and Koopman, data were acquired on single transfer for $C^{4+} + Ar$ and $C^{4+} + Ne$. Comparisons are presented in Table II. The average discrepancy is a factor of 3.2 and is independent of target.

The results of Goldhar, Mariella, and Javan[13] for $C^{4+} + He$ were obtained with a pulsed ion source from a CO_2 laser incident on a graphite target. These results are less definitively reported but are shown on Fig. 3 at the estimated velocity near 1×10^7 cm/sec. For double transfer they agree with calculation, while for single transfer they add further confusion.

The role of metastable ions in the incident beam has not been evaluated in any of the experiments. Since the energy required to produce metastable C^{4+} is nearly the same as that required to produce C^{5+}, it is reasonable that the metastable fraction will be of the same order as the ratio of C^{5+}/C^{4+} from the source. For the present experiment, this ratio is about 10^{-3}; for Zwally and Koopman, it is 10^{-2} or less; and for Goldhar *et al.*, it may be 10^{-1} or larger. The assumption that these fractions account for differences in observed cross sections by metastable content requires that single-electron capture by ions initially in the metastable states be at least 100 times larger than by ions in the ground state. There is no evidence to support such a conjecture, and in fact there is evidence that at higher velocities[14] capture by C^{4+} metastables in He is of about the same magnitude as capture by ground-state ions. Thus, it is doubtful that incident meta-

TABLE II. Values of σ_{43} in units of 10^{-16} cm^2 for C^{4+} on helium, neon, and argon from Zwally and Koopman (ZK), Ref. 9, and from present measurements (C).

Velocity		He			Ne			Ar	
10^7 cm/sec	ZK	C	Ratio	ZK	C	Ratio	ZK	C	Ratio
6.06	5.1	1.68	3.0	7.1			80	29.6	2.7
7.22	7.0	2.88	2.4	10.9	3.06	3.6	93		
8.37		3.20		15.2	4.08	3.7	110	28.2	3.9
9.04		3.86			4.65			27.9	

stables affect the observed cross sections.

At velocities higher than those used here, the cross sections are expected to decrease and the quasimolecular model is no longer valid. For C^{4+} + He, Dmitriev *et al.*[14,15] obtain σ_{43} of about 2×10^{-16} cm^2 and σ_{42} of about 3×10^{-18} cm^2 at 4×10^8 cm/sec. For double capture, this difference from present results is notable, but since the velocity gap between experiments is large no inconsistency can be claimed.

B. B^{3+} + He

Figure 4 shows both theory and experiment for single and double electron transfer for B^{3+} + He. Theory,[7] present data, and experiment of Zwally and Cable[10] agree to roughly 20% (well within combined uncertainties) for σ_{32}. Extrapolation of these data to the higher-energy experiments of Dmitriev *et al.*[13,14] is not unreasonable. The theoretical results of Shipsey *et al.*[7] show the calculated contributions of transfer to $B(2p)^{2+}$ and $B(2s)^{2+}$ states as well as the total single transfer. Figure 5 (from Ref. 7) shows their *ab initio* adiabatic potential curves, and illustrates that these two states are the only ones which lie below the initial state and have avoided crossings. The simplicity of the potential curves allows accurate calculation. Estimates of the potential curves ending on $B(3s)^{2+}$ + $He(1s)^+$ and $B(2s^2)^+$ + He^{2+} states are presented on Fig. 5 to show positions of the next-closest potential curves to initial state and to illustrate the double transfer case.

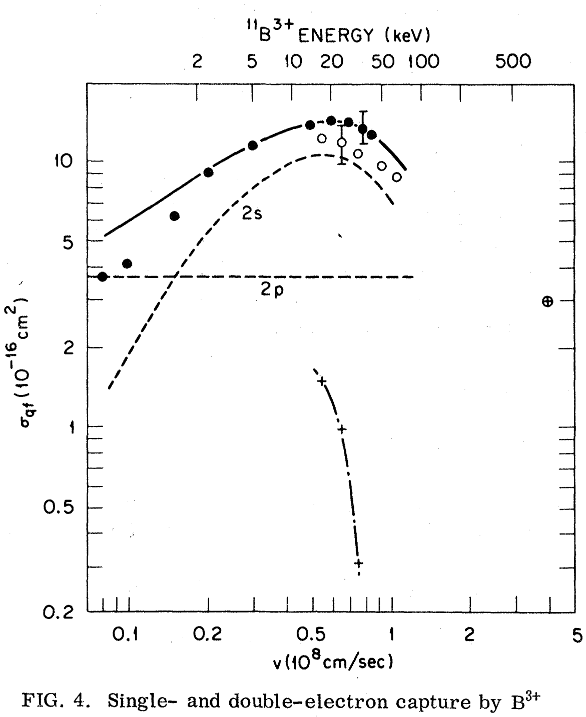

FIG. 4. Single- and double-electron capture by B^{3+} incident on He. Open circles, present data for single capture σ_{32}; crosses, present data for double capture σ_{31}; solid circles, σ_{32} from Ref. 10; circled cross, σ_{32} from Ref. 14; solid curve, calculation of total capture σ_{32} from Ref. 7; dashed curves, calculation of single capture, into $B^{2+}(2s)$ and $B^{2+}(2p)$ states.

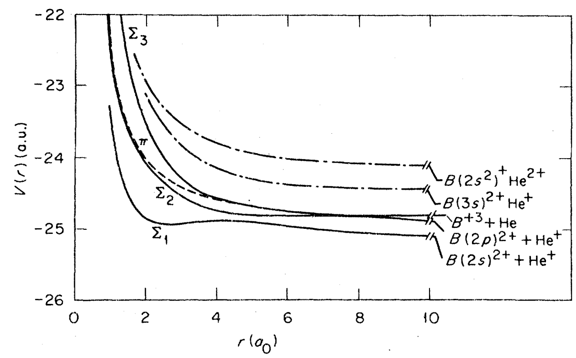

FIG. 5. Adiabatic potential curves for $(B-He)^{3+}$ from Ref. 7. Dot-dashed lines are estimated curves added for discussion.

The three data points in Fig. 4 for σ_{31} transfer of B^{3+} + He exhibit striking behavior. Since there is no direct curve crossing favorable to double capture, a small cross section is expected, as observed at 0.76×10^7 cm/sec (at the higher velocities the cross section was too small to measure). Nevertheless, by 0.55×10^8 cm/sec, the cross section has risen to 1.5×10^{-16} cm^2. Possibly a two-step transfer is responsible for the abrupt rise in this cross section. With this mechanism, adiabatic single transfer from the initial Σ_1 molecular state to the Σ_3 state at the avoided crossing near $r=7.5a_0$ is followed by transfer to the double capture final state at the strong repulsion near $r=2a_0$. The calculated cross section at the outer crossing is the $2p$ dashed curve of Fig. 4. Since this cross section is nearly constant with changing velocity, the observed variation of σ_{31} should result from the dynamics of the subsequent inner crossing. If the mechanism is valid, the $B(3s)^{2+}$ final state could be populated in a similar fashion.

Zwally and Cable present some data on the role of metastables in the incident beam of their B^{3+} + He experiment. From direct measurements, they estimated a metastable content of 3.7%, which resulted in a 2% correction to their cross section data. In the present experiment, the assumption that metastable B^{3+} would be about equal to B^{4+} population in the source implies a metastable component of the order of 1% in the B^{3+} beam. In other investigations[16] with our ion source, an attempt was made to measure electron impact ionization of B^{3+} in a crossed-beam experiment. Electrons of 61 eV or more could ionize metastable $B^{3+}(1s2s)\,^3S$; ionization of ground state $B^{3+}(1s^2)\ ^1S$ requires electron energy above 260 eV. Assuming that near threshold the ionization cross sections scale roughly as the inverse square of the ionization energy, we obtain a ratio of ionization out of the metastable to that out of the ground state $\sigma_m^I/\sigma_g^I \simeq 18$, indicating that the electron impact ionization experiment is a sensitive method for observing the presence of metastables. The first ionization measurements on our B^{3+} beam gave poor results, but allowed a crude estimate of metastable content. The ratio of apparent cross sections below threshold for ground-state ionization to that above was

$$\sigma^I(140\ \text{eV})/\sigma^I(480\ \text{eV}) = 0.29(\pm 0.15)$$

with $\sigma^I(140\ \text{eV}) \propto \sigma_m^I N_m/(N_g+N_m)$ and $\sigma^I(480\ \text{eV}) \propto (\sigma_m^I N_m + \sigma_g^I N_g)/(N_g+N_m)$, where N_g and N_m are the numbers of ground-state and metastable ions incident. Assuming $\sigma_m^I = 18\ \sigma_g^I$ as stated, one obtains $N_m/N_g \simeq 0.023\ (\pm 0.014)$, where the uncertainty is only the standard deviation of the measured cross-section ratio. This estimate is of the order expected, and suggests that metastable content in the present experiment is similar to that in the Zwally and Cable experiment; however, no correction is applied to the present cross sections.

C. N^{5+} and O^{6+} + He

Results for single- and double-electron capture by N^{5+} and O^{6+} incident on He are shown in Fig. 6. There are no other data or calculations for these cross sections below 2×10^8 cm/sec relative velocity. Measurements at higher velocities[14,15] begin at 4×10^8 cm/sec, with N^{5+} single-capture cross section about 5.5×10^{-16} cm^2 — a reasonable extrapolation from present data. The present single-electron-transfer cross sections are constant at

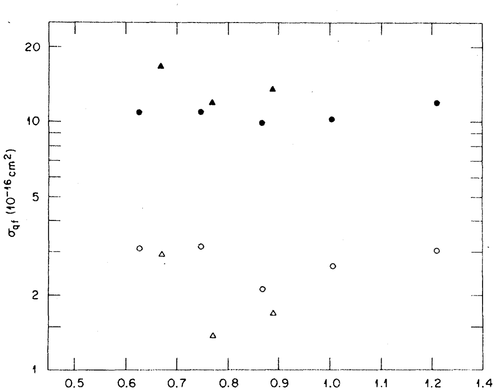

FIG. 6. Single- and double-electron capture for N^{5+} and O^{6+} incident on He. Solid circles—σ_{54} for N^{5+}; open triangles—σ_{65} for O^{6+}; open circles—σ_{53} for N^{5+}; open triangles—σ_{64} for O^{6+}.

about the same value as the maximum of σ_{32} for B^{3+} + He. The σ_{53} and σ_{64} are also roughly constant and about the same magnitude as σ_{42} for C^{4+} + He. For the velocities observed, these qualitative features seem typical of electron transfer between multicharged ions and neutrals when many molecular curve crossings become important. Figure 7 gives some of the diabatic curves for N^{5+} + He where the Coulomb repulsion of charge transferred final states is represented by $V = q_1 q_2/r$ and the long-range attraction of the initial state is represented by $V = -\frac{1}{2}\alpha(\text{He})q^2/r^4$, with α(He), the polarizability of helium, taken to be $1.37a_0^3$. All of the single transfer states lying below the initial state at infinite separation are represented except that final states with excited He^+ are not included, and only one of the N^{4+} ($n=3$) states is fully shown. Only a representative sample of the double-transfer final states is given. Detailed calculation is formidable, but some insight is available even from such a simple set of curves. The crossings at $r = 7$ to $8a_0$ leading to N^{4+} ($n=3$), suggest that population of these excited states is likely to be prominent, especially at velocities of the order of 1×10^7 cm/sec. As more states become involved, the absorbing sphere model of Olson and Salop[17] becomes appropriate. Their calculation assumes a large number of available final states and uses coupling matrix elements at the avoided crossings, which have been parametrized according to incident charge q and crossing distance r. Approximate cross sections are then obtained by an extension of the Landau-Zener approach. For single-electron transfer, present cases may not really have enough crossings to meet the assumptions of this model. The method has been applied for generalized ions incident on He at a velocity of 5.4×10^7 cm/sec giving 37×10^{-16} cm^2 for $q=5$ and 42×10^{-16} cm^2 for $q=6$. These values are about twice the present data.

The diabatic curves for O^{6+} + He are very similar to those for N^{5+}. For O^{6+}, the final states of O^{5+} (n

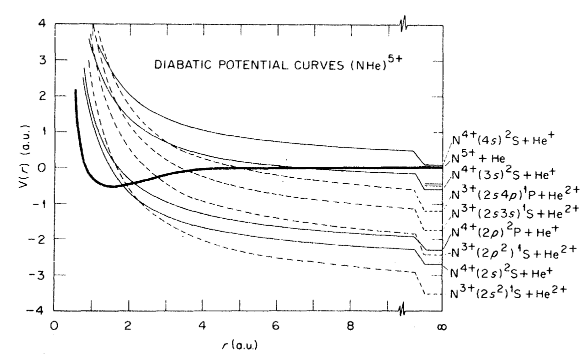

FIG. 7. Approximate diabatic potential curves for $(N\text{-He})^{5+}$.

= 3) cross the initial state near $r = 4$ to $5a_0$, a little closer than the N^{5+} case. Only a few O^{6+} data points were obtained, but behavior similar to N^{5+} is expected.

Double capture σ_{64} is favored to highly excited states; e.g., the $O^{4+}(2s4s)\,{}^1S + He^{2+}$ final state crosses the initial state about $2.6a_0$, where double capture may be fairly strong (as for σ_{42} for C^{4+}). For even more highly charged ions, double capture may be most favored to doubly excited states, which could then relax either by radiation or by autoionization appearing as single transfer plus a free electron.

CONCLUSIONS

Electron transfer between He-like multicharged ions and He has received significant attention as a system for which both calculation and experiment are accessible. For B^{3+} and C^{4+}, the molecular potential avoided crossings are sufficiently few and distinct that individual crossings affect the electron transfer cross sections, as revealed by variation of the cross sections with velocity and dominance of double transfer. Another interesting manifestation of selective curve crossing which remains untested is the population of particular excited states by electron transfer. Taken with other data,[1,18-20] the trend appears that for incident charge states $q < +4$, there are enough potential curve crossings favoring electron transfer between multicharged ions and neutrals to make the cross sections fairly constant, at least in the 10^7-cm/sec velocity region. The general decrease of these cross sections in the 10^8-cm/sec region is documented,[14,15,20,21] but there are no data for velocities below 10^7 cm/sec. The often-used scaling of cross sections with incident charge q does not appear to be appropriate for the present data.

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