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Single-Charge Transfer between He⁺⁺ and He, Ne, and Ar: Observed Selection Rule for Exit Channels*

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Single-charge-transfer channels in the systems He^{++} -He, -Ne, -Ar are observed for He^{++} energies 0.2–1.0 keV and $\text{He}^{+(*)}$ exit angles 0°–15°. ($\text{He}^{+(*)}$ denotes either the ground state or an excited state.) The ground-state channels have cross sections below the present 10^{-20} -cm² detection limit. Outgoing channels are found to be favored when their separated ions can originate with the same molecular symmetry as the incoming channel.

He⁺⁺ as a probe in ion-atom scattering, particularly charge transfer, provides many of the advantages of H⁺, viz., an incoming bare nucleus, a guaranteed ground-state beam, and a hydrogenic single-electron-attachment spectrum. It also has several useful distinctions: The singleattachment level spacings are 4 times as large as for H⁺, the molecular curves of single-chargetransfer channels are uniformly repulsive at large separations, and having both incoming (He⁺⁺) and outgoing (He⁺) species charged makes energy analysis and particle detection straightforward.

Figure 1 shows the charge-transfer channels originating on He⁺⁺-He, -Ne, and -Ar collisions, and indicates the observed channels. Zero energy here corresponds to the incoming channels, and endothermicity or exothermicity (Q_e) is the ordinate. A represents any one of the He, Ne, Ar targets; A^+ , the ground-state ion only; A^{+*} , an excited state only; $A^{+(*)}$, either the ground state or an excited state; and the form W(X,Y)Zsymbolizes projectile in (target in, target out) projectile out.

In a previous study of He⁺⁺-He, -Ne, and -Ar, Latypov, Flaks, and Shaporenko¹ collected the slow target ions, and so did not separate ionization and charge transfer. We have isolated individual channels by observing the kinetic energy and exit angle of the emerging projectile.² The present emphasis of single charge transfer should *not* be taken as implying that the double-chargetransfer cross sections are negligible.

For incident energies in the range 0.2-1 keV and for exit angles $0^{\circ}-15^{\circ}$, we have observed the



FIG. 1. Energy level diagrams, relative to incoming systems, showing the charge-transfer channels $He^{++}(A, A^{+(*)})He^{+(*)}$. (a) $He^{++}(He, He^{+(*)})He^{+}$; (b) $He^{++}(He, He^{+(*)})He^{+(*)}$; (d) $He^{++}(He, He^{+(*)})He^{+(*)}$; (d) $He^{++}(Ne, Ne^{+(*)})He^{+(*)}$; (d) $He^{++}(Ne, Ne^{+(*)})He^{+(*)}$; (e) $He^{++}(Ne, Ne^{+(*)})He^{+(n-2)}$; (f) $He^{++}(Ar, Ar^{+(*)})He^{+(*)}$; (g) $He^{++}(Ar, Ar^{+(*)})He^{+}$; (h) $He^{++}(Ar, Ar^{+(*)})He^{+(n-2)}$. Solid dots show observed channels.

channel spectra $\operatorname{He}^{++}(A, A^{+(*)})\operatorname{He}^{+(*)}$. The groundstate exit channels $\operatorname{He}^{++}(A, A^{+})\operatorname{He}^{+}$ are all below our 10⁻²⁰-cm² detection limit. At laboratory angles $\leq 3^{\circ}$, one channel, He⁺⁺(A, A^{+*})He⁺, dominates, where A^{+*} is He^{+*}(n = 2), or Ne^{+*}($2s2p^{6}$ -²S), or Ar^{+**} (autoionizing). At angles $\simeq 3^{\circ}-10^{\circ}$, several identifiable single-excitation channels appear, first with A^{+*} , then He^{+*}. At $\geq 10^{\circ}$, many unresolved multiple-excitation channels, $\operatorname{He}^{++}(A, A^{+*})\operatorname{He}^{+*}$, are seen. At a larger incident energy any given channel opens at a smaller angle. Representative data for the He, Ne, and Ar systems are shown in Figs. 2, 3, and 4. Each curve has been separately normalized: in this Letter we confine our discussion to our evidence for a stronger selection rule than the simple minimization of Q_e in determining chargetransfer exit channels.

Our apparatus consists of a mass-selected ion beam, a target gas cell, and rotatable collimation slits and 127° electrostatic energy analyzer. Detection is by a Channeltron multiplier operated as a current amplifier. Scattered current versus ion energy is recorded directly, or integrated for periods of 15-30 sec, as appropriate for the signal level. H_2^+ beam contamination is about 4%, but does not constitute a significant error source. The slit widths give resolutions of 0.5° (full width at half-maximum) and a $\Delta (Tq^{-1})/Tq^{-1}$ of 1%, where T is the particle energy and q the charge state.

Except for small kinematic corrections (whose leading terms depend quadratically on the scattering angle and Q_e) all of Q_e appears in the kinetic energy of the outgoing projectile. The chargetransfer peaks appear around twice the analyzer voltage, corresponding to the direct beam (or the elastic He⁺⁺ peak). Q_e is measured by the energy excess or defect, and each channel is identified by its location on Fig. 1. Any additive



FIG. 2. Representative data showing relative cross sections for channels $\text{He}^{++}(\text{He}, \text{He}^{+(*)})\text{He}^{+(*)}$ at various exit angles and incoming energies. For each energy and angle the data have been arbitrarily normalized.



FIG. 3. Same as Fig. 2, for $He^{++}(Ne, Ne^{+(*)})He^{+(*)}$ channels.

error in the analyzer-voltage measurement (such as a contact potential) produces a constant offset in all measured values of Q_e . We observe an offset of about 0.25 V in the He and Ne data (well within the expected range of contact potentials), and we subtract it from all our results. We also use this correction with the Ar data, in a range of Q_e where no spectroscopic data are available for comparison.

At our energy resolution the following points are pertinent: (1) The He II fine-structure levels are effectively degenerate; (2) He⁺(n = 2) can be separated from the He⁺ ground state and He⁺(n = 3), but for $n \ge 3$ all *n* levels are blended; (3) the Ne⁺* levels bunch into blocks $\simeq 1$ eV wide and 3– 4 eV apart, allowing resolution of the blocks but not individual levels within each block; (4) the Ar⁺* levels, with the exception of $3s 3p^{62}S$, are essentially continuous.³

Each ground-state channel is allowed by the "selection rule" given by $Hasted^4$ and confirmed



FIG. 4. Same as Fig. 2, for $He^{++}(Ar, Ar^{+(*)})He^{+(*)}$ channels.

by our Ne data: The separated ions of an allowed channel are able to combine, by the Wigner-Witmer rules,⁵ into the same molecular configuration as the incoming channel.⁶ However, the ground-state channels are all highly exothermic, by 29.62 eV (He), 32.84 eV (Ne), and 38.63 eV (Ar), probably implying small cross sections because curve crossings with the incident channel occur only high on the repulsive-core part of the molecular potential curves.

The He data (Fig. 2) cannot test this selection rule. In a hydrogenic system the total orbital angular momentum L fixes the parity, so any two terms satisfy the rule. Five He₂⁺⁺ curves have been calculated.⁷ He⁺(ls) + He⁺(ls) \rightarrow ¹ Σ_{g} ⁺ and ${}^{3}\Sigma_{u}$, He⁺⁺+He(1s²) - $({}^{1}\Sigma_{g})^{+}$ and ${}^{1}\Sigma_{u}$, and $\operatorname{He}^+(1s) + \operatorname{He}^+(2s) \to {}^{3}\Sigma_{g}^+$. These curves indicate that a crossing leading to $He^{++}(He, He^{+}(1s))He^{+}(1s)$ can occur, as anticipated, only for small impact parameters, implying a correspondingly small cross section. By contrast, the ${}^{1}\Sigma_{u}{}^{+}$ and $({}^{1}\Sigma_{g}{}^{+})*$ curves originating on the incoming He⁺⁺-He channel cross the ${}^{3}\Sigma_{g}^{+}$ + He⁺(1s)+He⁺(2s) curve about 10 eV above the separated atom level of the ${}^{s}\Sigma_{g}^{+}$ curves, and we would judge a transition likely. Unfortunately the singlet - triplet transition is forbidden⁴; we expect the He⁺⁺(He, He⁺)-He⁺(n = 2) channel to exit on the ${}^{1}\Sigma_{ug}^{+}$ + He⁺(1s) +He⁺(2s or 2p) curves, and these curves have not, to our knowledge, been calculated. Assuming that the steep rise of the ${}^{1}\Sigma_{u,g}^{+}$ curves, like that of the ${}^{3}\Sigma_{u,g}^{+}$ curves, begins at a significantly smaller internuclear separation than the He⁺⁺ +He curves, we would also expect the He⁺⁺(He, He^+) $He^{+*}(n=2)$ channel to be relatively likely.

The Ne data (Fig. 3) clearly demonstrate the selection rule. No potential curves for He⁺⁺-Ne are available, so no useful discussion in terms of curve crossings is possible. Table I gives the measured separations between the four peak centers of Fig. 3(d), and compares these separations with those obtained by allowing the outgoing channels He^{++} (Ne, Ne^{+*}) He^{+} with (1) all Ne^{+*} terms, (2) only the Ne^{+*} terms which satisfy the selection rule ${}^{1}\Sigma^{+} - {}^{1}\Sigma^{+}$, and (3) only Ne^{+*}(${}^{2}S$) terms. Case (3) corresponds to no coupling between nuclear translational and electron orbital angular momentum, while case (2) allows the exchange of angular momentum between nuclear translation and the electronic configurations of the separated ions, so long as no change of intermediate molecular symmetry occurs. While questions of appropriate weighting have clearly been inadequately handled, the table shows that

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TABLE I. Measured peak spacings [from Fig. 3(d)] compared with spacings calculated by permitting outgoing channels: (1) with no restriction, (2) restricted to terms satisfying the ${}^{1}\Sigma^{+} - {}^{1}\Sigma^{+}$ and Wigner-Witmer rules, (3) restricted to S terms.

Block location [Above Ne ⁺ (2 p ⁵) ² P ⁰]	Measured	(1)	(2)	(3)
	spacing	All	W-W	S
	(eV)	states	states	states
26.90 to 27.85 30.51 to 31.52 34.01 to 35.19 37.47 to 38.30	4.25 ± 0.15 3.25 ± 0.15 3.50 ± 0.15	3.567 3.745 3.070 $\frac{1}{3}[\sum (\text{the} 0.32)]$	4.003 3.401 3.535 ory-measurer 0.10	$\begin{array}{r} 4.443\\ 2.947\\ 4.010\\ ment)^2]^{1/2}\\ 0.21\end{array}$

case (2) provides the best agreement with the data.

For He⁺⁺-Ar (Fig. 4) we have neither potential curves, nor assigned energy levels where the charge-exchange peaks occur, above the Ar II ionization limit. Metastable autoionizing Ar II states at about 0.3 and 1.8 eV into the continuum have been reported,⁸ but we cannot say with certainty that these correspond to the levels we observe.

In conclusion, we have observed and identified individual charge-transfer channels for He⁺⁺-He, He⁺⁺-Ne, and He⁺⁺-Ar, and shown the gross features of their relative cross sections versus incoming energy and outgoing angle. We are modifying our apparatus to improve resolution and sensitivity, and to determine absolute cross sections. We hope these experiments will stimulate the calculation of potential curves for these systems.

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*Work sponsored by the U.S. Air Force under Contract No. F33615-69-C-1048. ¹Z. Z. Latypov, I. P. Flaks, and A. A. Shaporenko, Zh. Eksp. Teor. Fiz. <u>57</u>, 50, 1987 (1969) [Sov. Phys. JETP <u>30</u>, 29, 1076 (1970)].

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⁵G. Herzberg, *The Spectra of Diatomic Molecules* (Van Nostrand, New York, 1950), 2nd ed., pp. 318-319. ⁶He⁺⁺ plus the ¹S₀ target combine as ¹Σ⁺. The outgoing ground-state channels, He⁺(²S) and He⁺(²S), Ne⁺(²P⁰), or Ar⁺(²P⁰), can each yield a ¹Σ⁺ configuration. For He both ungerade and gerade terms occur.

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