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¹⁰I assume that two-photon decays, which proceed at the rate $A_{2\gamma} \approx 8.2Z^6 \text{ sec}^{-1}$, are not detected.

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Symmetric Resonance Double Charge Transfer in $\text{Kr}^{++} + \text{Kr}$ and $\text{Xe}^{++} + \text{Xe}$ Systems

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Cross sections of processes $\text{Kr}^{++} + \text{Kr} \rightarrow \text{Kr} + \text{Kr}^{++}$ and $\text{Xe}^{++} + \text{Xe} \rightarrow \text{Xe} + \text{Xe}^{++}$ were measured by the injected-ion drift-tube technique from 0.04 to 20 eV. For both cases, the cross section below 1 eV coincides with the orbiting cross section with a charge-transfer probability $\frac{1}{2}$. Above 1 eV, the energy dependence of the cross section is like that for single charge transfer. Mobilities of Kr^{++} and Xe^{++} in He are presented also.

Although a number of studies have been made on symmetric resonance single-charge-transfer processes, relatively little work has been reported on symmetric resonance double charge transfer. Direct measurements of the cross section are completely nonexistent in the sub-electron-volt region. Here, we report the results of cross-section measurement of the processes



and



by the injected-ion drift-tube technique¹ in the energy range from 0.04 to 20 eV.

The details of the apparatus used in this work have been described previously.^{2,3} The doubly charged ions produced in a conventional ion source of electron-impact type are selected by a magnetic field of sector type, and injected into the drift tube with an energy of 20 eV. The drift tube is filled with He gas at about 1 Torr. The He gas is used as buffer gas. The ions injected are slowed down quickly to thermal energy by collisions with He atoms. The ions drift toward the end of the drift tube under the action of a uniform electric field applied along the axis of the tube.

The drift velocity of the ions is measured by the gate-pulse technique.^{2,3} The mean energy of the drifting ions, which is given as a function of the drift velocity, can be controlled over a wide range from room temperature by varying the ratio of the electric field strength X to the number density of gas N , X/N . A very small amount of natural Kr or Xe gas is mixed with the He buffer to the extent of about 0.05%. During the drift, some of the primary ions collide with the reactant atoms, and produce secondary ions by double electron capture. The product ions drift as well as the primary ions. They are analyzed with a sampling mass spectrometer attached on the end of the drift tube. From the intensity ratio of the secondary ions to the primary ions, I_2/I_1 , the cross section is determined.

The mean energy of drifting ions is given by Wannier's formula,⁴

$$E = \frac{1}{2}(m_i + m_b)v_d^2 + \frac{3}{2}k_B T_G, \quad (3)$$

where m_i is the mass of the ions, m_b is the mass of the buffer-gas molecules (He), v_d is the drift velocity, and T_G is the temperature of the buffer gas. Since a drift velocity is one of the fundamental quantities of gaseous electronics, and is often given in the form of a reduced mobility, the re-

duced mobilities obtained for Kr^{++} and Xe^{++} in He are shown in Fig. 1, together with those for Kr^+ and Xe^+ . There are big differences between the mobilities of singly and doubly charged ions in He. They provide a very interesting problem, but it is not our present purpose to get into this problem further. It will be discussed elsewhere.

The average cross section is given by

$$\sigma = \frac{v_d}{v_r N l} \ln \left(\frac{I_2}{I_1} + 1 \right), \quad (4)$$

where N is the number density of reactant atoms, and l is the length of the drift tube. The mean relative velocity v_r is given by

$$v_r = (2E/m_i + v_t^2)^{1/2}, \quad (5)$$

where v_t is the thermal velocity of reactant atoms. Since the secondary ions partake of the natural isotope abundance ratio, the ratio I_2/I_1 must be replaced by

$$\frac{I_2}{I_1} = \frac{I_2'}{a_2 I_1' - a_1 I_2'}, \quad (6)$$

where I_1' is the peak height at the primary ion mass in the sampling mass spectra, and I_2' is that at any of the isotopes. The constants a_1 and a_2 are the natural abundance ratios corresponding to each of the isotopes. These procedures are essentially the same as those for symmetric resonance single charge transfer, and the readers should refer to Ref. (2) for further details. In this work, $^{84}\text{Kr}^{++}$ and $^{129}\text{Xe}^{++}$ are used as primary ions, and $^{86}\text{Kr}^{++}$ and $^{136}\text{Xe}^{++}$ are chosen for secondary ions for reasons of intensity and mass resolution.

It is fortunate that the recombination energies

$R(\text{Kr}^{++} - \text{Kr}^+)$ and $R(\text{Xe}^{++} - \text{Xe}^+)$, which are 24.56 and 21.2 eV, respectively, are smaller than the ionization potential of He, which is 24.58 eV. Therefore, a process like $\text{Kr}^{++} + \text{He} \rightarrow \text{Kr}^+ + \text{He}^+$ rarely occurs. A process like $\text{Kr}^{++} + \text{Kr} \rightarrow \text{Kr}^+ + \text{Kr}^+$ may occur, but the cross section will be much smaller than that for a resonance process. Although small peaks of Kr^+ , Xe^+ , and He^+ are seen in the sampling mass spectra, they are thought to be produced by fast neutrals which are produced along the primary ion beam path and get into the drift tube. These ions can be distinguished from true signals by deflecting the primary ion beam with a deflector situated in front of the drift tube.⁵ They do not interfere with the cross-section measurement of resonance charge transfer anyway.

In the present case, the masses of the primary and secondary ions are almost the same, and so are their reduced masses in He. Therefore there are no problems, which might otherwise arise in some cases, due to the difference in the diffusion in the drift tube or the detection efficiency of the sampling mass spectrometer.

Experimental errors mainly arise from the measurement of I_2/I_1 and the determination of the reactant gas density N . The statistical error in the measurement of I_2/I_1 is about 10%. The uncertainty in the determination of N , which arises from the correction factor for the "concentration effect"⁶ of the reactant gas in the drift tube, is estimated to be 10%. The overall uncertainty of the cross-section measurement is estimated to be $\pm 20\%$. For relative values of cross section, $\sigma(\text{double charge transfer})/\sigma(\text{single charge transfer})$, the uncertainty may be smaller than this.

A cross section obtained by the drift-tube technique is a sort of "average cross section." The energy distributions of drifting ions have been studied theoretically.⁷ When the buffer gas is He, the energy distribution is supposed to be a Maxwellian at low energy, and a little narrower than the Maxwellian at high energy.⁷ The energy distribution may affect the average cross section, of course. However, the average cross section would not be much different from the true cross section when the energy dependence of the cross section is weak.

In Fig. 2, the cross sections obtained for double- and single-charge-transfer reactions are shown. The abscissa is the collision energy in the center-of-mass system, which is given by $\frac{1}{2}\mu v_r^2$, where μ is the reduced mass of the ion with the atom.

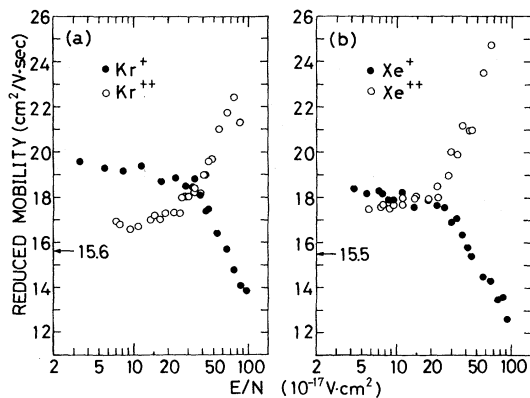


FIG. 1. The reduced mobilities obtained in He. Arrows indicate the zero-field limits of the Langevin theory.

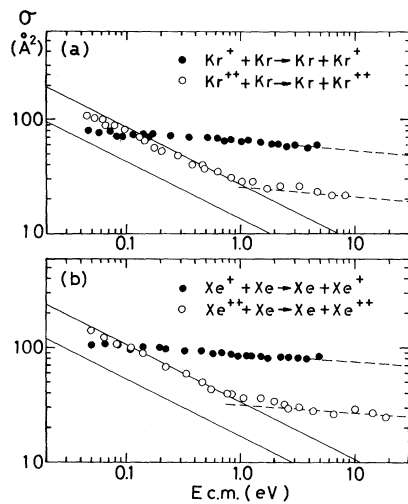


FIG. 2. The cross sections obtained for resonance-charge-transfer reactions. The solid lines indicate the σ_{orb} given by Eq. (7) taking q as 1 or 2. The dashed lines are drawn to fit the experimental points and to be parallel with each other.

General features of the experimental results are quite similar for Kr and Xe. The double-charge-transfer cross section appears to be divided into two parts at about 1 eV. Above 1 eV, the cross-section curve decreases slowly with the increase of energy and tends to parallel that of single charge transfer. Below 1 eV, its slope is steeper and the energy dependence is almost $E^{-1/2}$. It is striking that the double-charge-transfer cross section exceeds that of single charge transfer even at room temperature.

The solid lines in Fig. 2 indicate the half of the orbiting cross section due to the polarization force:

$$\sigma_{orb} = \frac{1}{2} \frac{2\pi q e}{v_r} \left(\frac{\alpha}{\mu} \right)^{1/2}, \quad (7)$$

where q is the number of charge of the ion, e is the electronic charge, and α is the polarizability of the atom. The factor of $\frac{1}{2}$ is taken to account for the charge-transfer probability. Rapp and Francis⁸ predicted for resonance single

charge transfer that the cross section will be σ_{orb} at very low energy. It has not yet been confirmed experimentally, however, because the transition from the linear-trajectory model to the orbiting model occurs a little below room temperature for ordinary resonance single charge transfer.

The experimental cross sections obtained for double charge transfer below 1 eV coincide with σ_{orb} . This is the first direct evidence that in low energy, resonance double-charge-transfer cross section is given by the orbiting model taking the charge-transfer probability as $\frac{1}{2}$.

Along the parallel portion above 1 eV, the ratio of the cross section of double charge transfer to single charge transfer is determined to be 0.40 for Kr and 0.36 for Xe. It should be noted that the ratio is close to the ratio of the double ionization potential to the single ionization potential of the atoms, which is 0.36 for both Kr and Xe.

It would be very interesting to know whether a resonance multiple charge transfer such as $A^{+q} + A \rightarrow A + A^{+q}$ takes place for $q \geq 3$. If it occurs with a large probability for large q , the orbiting mechanism will make its cross section very large in the low-energy region.

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