Charge Transfer between O^{2+} Ion and Helium at Electronvolt Energy

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The charge transfer rate coefficient for the reaction $O^{2+} + He \rightarrow O^+ + He^+$ was measured at energies about 2.5 eV (2×10^4 K) using a laser-induced-plasma ion source and ion storage. The rate coefficient is $1.12(0.14) \times 10^{-12}$ cm^3 s^{-1}, and disagrees with the available theoretical value. Our measured rate coefficient is also different from the value obtained by drift tubes at lower energies by at least an order of magnitude. The charge transfer rate coefficient for O^{2+} and the residual gas (H_2, H_2O, and CO) of the ultrahigh vacuum system is estimated to be $\sim 5 \times 10^{-9}$ cm^3 s^{-1}.

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The charge transfer reaction $O^{2+} + He \rightarrow O^+ + He^+$ has generated considerable interest as a potential mechanism for the destruction of O^{2+} ions, and the source for O^+ and He^+ ions in the terrestrial ionosphere, in supernovae, and in the upper atmospheres of Mars and Venus [1]. However, there are significant discrepancies between the results of theoretical quasimolecular calculations and the low energy measurements on the cross sections and rate coefficients at a wide range of energies [2–6]. The disagreement is particularly dramatic at energies below 1 eV/amu. Not only is the calculated rate coefficient different from the experimental values by at least a factor of 2 [1,7,8], but there is a 3 orders of magnitude difference between the only two measurements using drift tubes [7,9]. The reaction rate of this charge transfer process clearly needs further study to resolve these discrepancies.

The experimental technique recently used to measure the thermal-energy charge transfer rate coefficient of the Mo^{6+} ion and argon [10] is well suited to measurements at low energy. It uses a laser ablation ion source and ion storage [11]. With this technique, ions are produced directly from a solid target, and no source gas is required. Reaction between ions and source gas can be a potential source of systematic error. In addition, the mass selectivity of the ion trap enables us to store a specific ion group for study with no other concomitant ions present.

The facilities used in this study have been described earlier by Kwong *et al.* [11]. Minor modifications of the ion detection system were made to improve mass resolution and ion collection efficiency. The ion detection system includes a 0.3-m time-of-flight (TOF) mass spectrometer equipped with an electrostatic lens and a 1-in.-diam dual microchannel plate for better ion collection and higher gain. O^{2+} ions are produced by laser ablation of solid ferric oxide (Fe_2O_3) targets. Ferric oxide was chosen as the target material because of the distinct difference in the mass-to-charge ratio between low charge states of iron ions (Fe^+, $m/q = 56$, and Fe^{2+}, $m/q = 27$) and oxygen ions (O^+, $m/q = 16$, and O^{2+}, $m/q = 8$). We purposely used very low laser energy (0.3 mJ per laser pulse) for ion production to eliminate the formation of higher charge states of iron and oxygen. The ion trap parameters were chosen so that only O^{2+} ions are stored in the trap. Figure 1 shows the time-of-flight mass spectrum of the laser produced O^{2+} ions stored in the trap.

The experimental procedure has been discussed in detail elsewhere [10,11]. We shall only provide a brief summary. Ultrahigh purity helium gas was admitted to the vacuum chamber raising the pressure to a value measured with a calibrated ion gauge. The purity of helium gas was confirmed by a residual gas analyzer (Masstorr DX 100) installed in the vacuum chamber. O^{2+} ions were created by laser ablation and were cooled and stored in the radio frequency ion trap. At a delay time relative to the ablation event, two complementary push/pull extraction pulses (300 V, 1 μs wide) were applied to the end caps of the rf trap to empty all the stored ions into the 0.3-m TOF mass spectrometer. The time-of-flight mass spectrum was recorded by a transient digitizer for later analysis. The storage time, t, was scanned from its minimum value (~ 0.4 s) by increasing the delay time by a fixed increment Δt after each measurement until the ion signal intensity had dropped by one decade from its

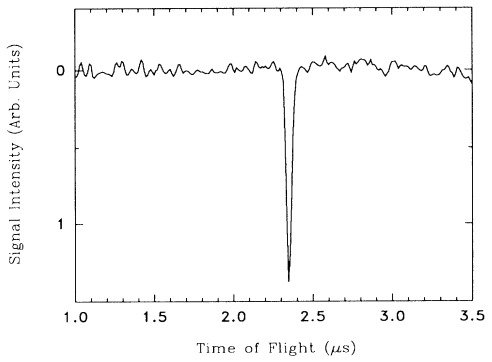

FIG. 1. A time-of-flight mass spectrum for laser (0.3 mJ per pulse) produced O^{2+} ions stored in the trap. Under the trap conditions (rf frequency $f = 1.4$ MHz, rf amplitude $V_0 = 380$ V, and dc bias $U_0 = 35$ V), O^{2+} is the only trapped ion. The TOF pulse is narrower than the extraction pulse sequence because of a time focusing effect.

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value at the shortest time delay. The storage time was then scanned in the opposite direction by decreasing the delay time by fixed Δt. About ten such cycles were used to obtain the data for analysis.

O^{2+} ions can be in a variety of excited electronic states immediately after they are produced by laser ablation. Rapid cascade from these excited electronic states through allowed transitions and collisional deexcitations of the excited states by the plasma electrons leave O^{2+} ions in their low lying metastable $2p^2\ ^1D$ (mean lifetime ~ 37 s) and $2p^2\ ^1S$ (mean lifetime ~ 0.55 s) states [12] and the $2p^2\ ^3P$ ground state during the early expansion phase into the vacuum. The ratio of the number of metastable state ions to ground state ions depends on the density and the temperature of the plasma electrons during the self-similar expansion in the vacuum chamber [13]. At a laser energy of 0.3 mJ (~ 0.05 mJ above the ablation threshold), the time of flight of the plasma electrons measured by a Langmuir probe mounted 7.9 cm away from the target surface revealed a maximum electron energy of 1.1×10^{-3} eV and an electron density of the order of 10^9 cm^{-3}. Using the empirical relationship of t^{-1} dependence of the electron temperature and t^{-3} dependence of the electron density obtained by Rumsby and Paul [13] with laser induced carbon plasmas, the electron temperature of the laser induced plasma near the target surface at the termination of a 20-ns ablation laser pulse is estimated to be 2×10^{-1} eV and the corresponding electron density is $\sim 10^{16}$ cm^{-3}. The plasma is highly collisional in the early phase of expansion [14]. A conservative estimate on the ratio of the metastable state population to the ground state population is of the order of 10^{-6} to 10^{-5}. As the plasma expands and cools, the population ratio will decrease further. Such a small number of metastable state O^{2+} ions has no effect on our measurement. The observed single exponential decay curve of the O^{2+} ion signal in Fig. 2 clearly confirms this.

The mean energy of the stored ions in an rf trap is known to be approximately one-tenth of the potential well depth qD [15,16]. Since the well depth for the rf trap was set at $qD = 24.5$ eV, the energy of the stored O^{2+} ions is about 2.5 eV.

A total of seven or more individual measurements were made for the collisional decay rate of O^{2+} ions at each He pressure, with each measurement corresponding to ten cycles of the storage time scans as described earlier. The decay rate for each He pressure was obtained by averaging the decay rates determined from each individual measurement at that pressure. Figure 2 is a plot of the ion signal intensities, summed data over all seven measurements, versus delay time, t, after laser ablation for several helium gas pressures. The scatter on the data points is due to the fluctuations of the ion signals. Each solid line is a least-squares fit of the data by a single exponential decay function. The slope of a fitted line in Fig. 2

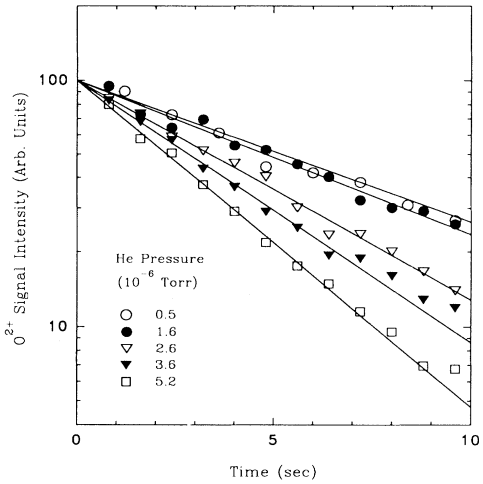

FIG. 2. The decay curves of O^{2+} ion signal vs storage time for different helium pressures.

gives the charge transfer rate at a given helium pressure. The statistical errors for the rates were estimated from the scatter of the results from separate measurements. The plot of the decay rates of the stored O^{2+} ions versus He pressure is shown in Fig. 3. The slope in the figure is obtained by the weighted least-squares fits by a linear function. The rate coefficient for O^{2+} and He obtained from the slope of Fig. 3 is $1.12(0.14) \times 10^{-12}$ cm^3 s^{-1}. The uncertainties presented are computed from the uncertainties of the helium density measurements ($\pm 8\%$) and the statistical uncertainties derived from the linear least-squares fit of the measured decay rates at different He pressures in Fig. 3. Table I summarizes the results of various calculations based on the quasimolecular model and experimental measurements. Our measured value is significantly different from the calculations of Dalgarno

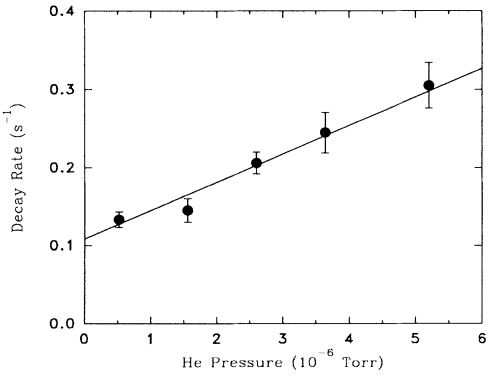

FIG. 3. O^{2+} decay rate vs helium pressure. Each error bar represents the statistical uncertainty of 1σ. The slope of the straight line fit gives the charge transfer rate coefficient of the O^{2+} ion with He, while the intercept at zero helium pressure gives the charge transfer rate of the O^{2+} ion with the residual gas in the vacuum chamber.

TABLE I. Summary of charge transfer rate coefficient, K, for O^{2+} and He at eV energies.

Reference	K ($cm^3\ s^{-1}$)	Temperature (K)	Method
[7]	$\leq 10^{-14}$	300	Experiment, drift tube
[8]	8×10^{-12}	300	Theory, quantal calculation
[9]	$3.5(1.5) \times 10^{-11}$	400	Experiment, drift tube
[6]	2×10^{-11}	400	Theory, distorted wave
[1]	10^{-14}	300	Theory, quantal calculation
	$4 \times 10^{-14} - 4 \times 10^{-13}$	1×10^3	
	$2 \times 10^{-11} - 1 \times 10^{-10}$	1×10^4	
[8]	1.0×10^{-10}	5×10^3	Theory, quantal calculation
	2.0×10^{-10}	1×10^4	
	3.9×10^{-10}	2×10^4	
	5.9×10^{-10}	3×10^4	
	8.9×10^{-10}	5×10^4	
This work	$1.12(0.14) \times 10^{-12}$	2×10^4 [a]	Experiment, ion trap

[a]Estimated from $\bar{E} = \frac{3}{2}kT$ with $\bar{E} \approx 2.5$ eV. Previous measurements in an rf trap indicate that the ion energy distribution is approximately thermal [17].

et al. [1], and the drift tube measurements by Johnsen and Biondi [9], and Howorka *et al.* [7] at lower energies.

The charge transfer rate coefficient of O^{2+} and the residual background gas (H_2, H_2O, and CO) of an ultrahigh vacuum system is also measured. The rate coefficient is $\sim 5 \times 10^{-9}$ $cm^3\ s^{-1}$ and was obtained from the intercept at zero helium pressure of the charge transfer rate in Fig. 3. This value is consistent with the rate coefficient obtained from the decay constant of the O^{2+} ions signal at the base pressure of 6×10^{-10} Torr in the vacuum chamber.

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