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STATUS REPORT OF GRANT NAGW-4279

TITLE: Laboratory Studies of Thermal Energy Charge
 Transfer of Silicon and Iron Ions in
 Astrophysical Plasmas

PRINCIPAL
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PERIOD: January 1, 1996 - December 31, 1996

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GRANT NUMBER: NAGW-4279

The laser ablation/ion storage facility at the UNLV Physics Department is dedicated to the study of atomic processes in low temperature plasmas. Our current program is directed to the study of charge transfer of multiply charged ions and neutrals that are of importance to astrophysics at energies less than 1 eV (about 10^4 K). Specifically, we measure the charge transfer rate coefficient of ions such as N^{2+} , Si^{3+} , Si^{4+} , with helium and Fe^{2+} with molecular and atomic hydrogen. All these ions are found in a variety of astrophysical plasmas. Their electron transfer reactions with neutral atoms can affect the ionization equilibrium of the plasma.

We have had a very productive year. We have completed several major tasks that include [A] the re-calibration of the ion gauge to measure the absolute particle densities of H_2 , He, N_2 , and CO for our current measurements; [B] the analysis of data for charge transfer reactions of N^{2+} ion and He, H_2 , N_2 , and CO; [C] measurement and data analysis of the charge transfer reaction of Si^{3+} and He; [D] charge transfer measurement of Fe^{2+} ion and H_2 ; and [E] redesigning and modification of the ion detection and data acquisition system for the low energy beam facility (reflection time of flight mass spectrometer) dedicated to the study of state select charge transfer. Details of each accomplishment are outlined in the following paragraphs.

A) The absolute calibration of the ion gauge to measure the particle densities of H_2 , He, N_2 , and CO was completed. The absolute particle densities of these neutral atoms and molecules are needed to determine their absolute rate coefficients. We did not find any significant change in the calibration of the ion gauge since our last calibration five years ago. We estimated that the uncertainty on the absolute density measurement is less than 8%.

B) We measured the charge transfer reactions of N^{2+} ion and He, H_2 , N_2 and CO. Results of these measurements will be reported in Ap.J. and Physical Review A in 1997. In the first paper, "Measurement of charge transfer rate coefficient between ground state N^{2+} ion and He at electron-volt energies" (Fang and Kwong, 1997a), our measurement confirmed the full quantal calculation of Sun et al. (1996). The measurement reported in the next paper, "Charge transfer between ground-state N^{2+} and H_2 , N_2 , and CO at electron-volt energies" (Fang and Kwong, 1997b), however, does not have any theoretical value for comparison. The lack of theoretical studies may be due, in part, to the complicated electronic, vibrational, and rotational structures of the molecules and the availability and the accuracy of the position and orientation sensitive potentials of the pseudo-molecule formed during the collision. Our paper is a step towards making these calculations possible. Furthermore, our paper also makes these new data available to the astronomical

community for plasma modeling.

C) We measured the charge transfer reaction of Si^{3+} and He. The results are reported in "Charge transfer between ground-state Si^{3+} and He at electron-volt energies" (Fang and Kwong, 1997c) which has been submitted to Ap.J. Silicon is commonly found in a variety of astrophysical plasmas. Their spectral lines have been observed in interstellar medium, circumstellar material, quasars, Seyfert galaxies, solar lower transition region and late-type stellar atmospheres and has been used for plasma diagnostic. However, the ionization structure of silicon ions can be modified by charge transfer process. Our result is the first and only measurement available at this low energy region. It is within 30% of the Landau Zener approximation of Butler and Dalgarno(1980) and a factor of three larger than the full quantal calculation of Honvault et al.(1995). Our current and previous findings (Kwong and Fang, 1993, Fang and Kwong, 1995) call to question some of the calculation results currently available.

D) We also completed a preliminary measurement on the charge transfer rate coefficient of $\text{Fe}^{2+} + \text{H}_2$. We estimated that the rate coefficient is less than $10^{-14} \text{ cm}^3\text{s}^{-1}$. This is an unexpectedly small rate since $(\text{FeH}_2)^{2+}$ has a complex atomic structure with numerous low lying energy states where charge transfer can occur. This measurement will be useful in the measurement of $\text{Fe}^{2+} + \text{H}$ where small amounts of H_2 are expected to be present in the atomic hydrogen beam.

E) We redesigned the low energy ion beam facility (reflection time-of-flight mass spectrometer) to eliminate the uncertainty on the measurement of charge transfer cross-sections created by the differential gain of the ion detector for ions at different charge state and energy. This differential detector gain is caused by the difference in the secondary electron emission efficiency of the detector surface when an ion of different charge state and/or energy strikes. We have eliminated this problem by adopting the ion counting technique using a new system which includes a Phillips Scientific fast pre-amplifier, a high speed Ortec multichannel analyzer with 2 ns per channel resolution and a Tektronix TDS-680B digital oscilloscope supported by a 200 MHz PC for data storage and analysis. This facility was designed and built by Mr. J.B. Wang as part of his Ph.D. project to study state select charge transfer. This facility complements the current laser plasma/ion storage facility by quantifying both the parent and the daughter ions of the reaction.

We have delayed measurements of Si^{4+} and He due to the rapid deterioration on the performance of the K*DP crystal used in the second harmonic (532 nm) generation. The second harmonic of the Nd:YAG has been used as an ablation source for the production of

multiply charged ions. We expect that the refurbished K*DP crystal be installed in the laser system by the end of November and we can begin the Si⁴⁺ He measurement before the end of this year.

In the coming year, we shall refine our measurement of the charge transfer rate coefficient of Fe²⁺ + H. Preparation for the measurement of the spatial density profile and the absolute density of atomic hydrogen beam using the calibration technique of Wise, Roberts and Haerberli (1993) is also underway. In addition, Mr. De Chen has joined the ion trap program this fall to study state select charge transfer as part of his doctoral research. His study is directly related to astrophysical plasmas since some of the photo emission from astrophysical plasmas can be related to electron capture by multiply charged ions into their excited states.

TABLE 1 **SUMMARY OF OUR MEASUREMENT RESULTS**

| ION | NEUTRAL | RATE COEFFICIENT | | TEMPERATURE (K) |
|------------------|----------------|------------------------------|--|--------------------|
| | | MEASUREMENT | CALCULATION (cm ³ s ⁻¹) | |
| N ²⁺ | He | 8.67(0.76)x10 ⁻¹¹ | 8x10 ⁻¹¹ [Sun et al.1996] | 3900 |
| | H ₂ | 3.38(0.35)x10 ⁻¹¹ | N/A | 2900 |
| | N ₂ | 2.10(0.18)x10 ⁻⁹ | N/A | 13000 |
| | CO | 3.37(0.29)x10 ⁻⁹ | N/A | 13000 |
| Si ³⁺ | He | 6.27(0.52)x10 ⁻¹⁰ | 4.5x10 ⁻¹⁰ [Butler & Dalgarno,1980] 2x10 ⁻¹⁰ [Honvault etal.1996] | 3900 |
| Fe ²⁺ | H ₂ | < 10 ⁻¹⁴ | N/A | 4000 |

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 Chem., 15, 53.
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BUDGET SUMMARY FOR YEAR 3 - NAGW-4279

TITEL:Laboratory Studies of Thermal Energy Charge Transfer of Silicon and Iron Ions in astrophysical Plasmas

| BUDGET ITEM | \$ |
|---------------------------|---------------|
| ----- | ----- |
| Senior Scientist 20% Time | 12,808 |
| Fringe Benefits | 368 |
| Travel | 1,025 |
| Other Direct Cost | 2,192 |
| ----- | ----- |
| Total Direct Cost | 16,393 |
| Indirect Cost @46.4% MTDC | 7,607 |
| ----- | ----- |
| Total Cost | <u>24,000</u> |

MEASUREMENT OF CHARGE TRANSFER RATE COEFFICIENT BETWEEN GROUND-STATE N^{2+} ION AND He AT ELECTRON-VOLT ENERGIES

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ABSTRACT

The charge transfer rate coefficient for the reaction $N^{2+}(2p^2P^o) + He \rightarrow$ products is measured by recording the time dependence of the N^{2+} ions stored in an ion trap. A cylindrical radio-frequency ion trap was used to store N^{2+} ions produced by laser ablation of a solid titanium nitride target. The decay of the ion signals was analyzed by single exponential least-squares fits to the data. The measured rate coefficient is $8.67(0.76) \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$. The N^{2+} ions were at a mean energy of 2.7 eV while He gas was at room temperature, corresponding to an equivalent temperature of $3.9 \times 10^3 \text{ K}$. The measured value is in good agreement with a recent calculation.

Subject headings: atomic data — atomic processes — methods: laboratory

1. INTRODUCTION

In the interstellar gas, multiply charged ions are formed and co-exist with neutral atoms of hydrogen and/or helium. Since the interstellar gas is not in thermodynamic equilibrium because of its low density (Steigman 1973, 1975), the distribution of elements among its various ionization stages is governed by several competing processes including charge transfer, dielectronic recombination, radiative recombination, electron impact ionization, and photoionization. Depending on the environment, charge transfer can be the dominant process that controls the states of ionization in astrophysical plasmas (Dalgarno 1985; Shields 1990; Dalgarno & Fox 1994). For a realistic modeling of these astrophysical plasmas, it is of paramount importance that the charge transfer rate coefficients be known accurately at the temperatures of these environments (Shull 1993).

In the past, the Landau-Zener model was used to estimate the charge transfer cross sections and rate coefficients. However, this technique may fail at low energies. Vigorous quantal calculations have been used to provide more accurate results. At present, there are few experimental tests of these calculated values and their accuracy. Our recent measurement of the charge transfer rate coefficient of O^{2+} and He (Kwong & Fang 1993; Fang & Kwong 1994) indicates that the measured rate coefficient is three orders of magnitude smaller than the full quantal calculations by Gargaud, Bacchus-Montabonel, & McCarroll (1993) and an order of magnitude smaller than the Landau-Zener calculation of Butler, Heil, & Dalgarno (1980).

Charge transfer of N^{2+} and He at eV energies can play an important role in stellar chemistry as a source of N^+ ions in nebulae (Dalgarno 1985; Sun et al. 1996). Three calculations have been made at this energy range. Calculations made by Nikitin & Reznikov (1913) and Lafyatis, Kirby, & Dalgarno (1993) were based on the Landau-Zener model while Sun et al. (1996) carried out a sophisticated multistate close-coupling quantal calculation. However, all these calculations disagree with the measurement of Sadilek et al. (1990) using beam scattering at eV energies. In this paper, we report a measurement of the total charge transfer rate coefficient of the reaction by using a laser ablation ion source and ion storage technique.

2. EXPERIMENTAL METHOD

The measurement of the ground-state thermal energy charge transfer rate coefficient between N^{2+} ion and helium is carried out by using the technique of laser-ablation ion source and ion storage that is uniquely developed in this laboratory and dedicated for low-energy charge transfer studies. We have described in detail this measurement technique in our previous publications (Kwong et al. 1990; Fang & Kwong 1994, 1995).

The N^{2+} ions are produced by laser ablation of solid titanium nitride (TiN) targets. Titanium nitride was chosen as the target material because of the distinct mass-to-charge ratio difference between N^{2+} ions ($m/q = 7$) and other low-charge-state ions produced from the target (N^+ , $m/q = 14$; Ti^+ , $m/q = 48$; Ti^{2+} , $m/q = 26$; Ti^{3+} , $m/q = 16$; etc.). The measurement of $Ti^{4+} + He$ will be reported elsewhere. We used the output of the second harmonic (532 nm) of a pulsed Nd:YAG laser as an ablation source. In this series of measurements, approximately 1 mJ of the second harmonic of the Nd:YAG laser was used. The power density was estimated to be about 10^8 W cm^{-2} with laser pulse duration of about 25 ns. The low power density of the ablation source eliminates the formation of higher charge states of titanium and nitrogen. The prominent feature of this method is that no source gas is used in ion production, therefore the complexities and uncertainties caused by the interaction between the ions and the source gas are totally eliminated. A cylindrical radio-frequency (rf) quadrupole ion trap serves as a mass-selective ion storage device. The trapping parameters (rf $f = 1.44 \text{ MHz}$, amplitude $V_0 = 350 \text{ V}$, and dc bias $U_0 = 29 \text{ V}$) were chosen to selectively store N^{2+} ions. The axial well depth was $D_z = 26.6 \text{ eV}$ and the radial well depth was $D_r = 35.6 \text{ eV}$ for N^{2+} ions. The low charge states of titanium ions and singly charged nitrogen ions were excluded from the trap. To detect the stored ions, two extraction pulses (+190 V and -200 V) were applied simultaneously to the upper and lower end caps of the trap, respectively. These push and pull extraction pulses empty all the stored ions into a 0.3 m time-of-flight (TOF) mass spectrometer where the signal was detected by a 1 inch (2.5 cm) diam dual microchannel plate. The TOF mass spectrum was recorded by a transient digitizer and stored in a computer for later analysis. The TOF signal was used to identify the ion species while the signal intensity served to measure the population of the ions stored in the trap prior

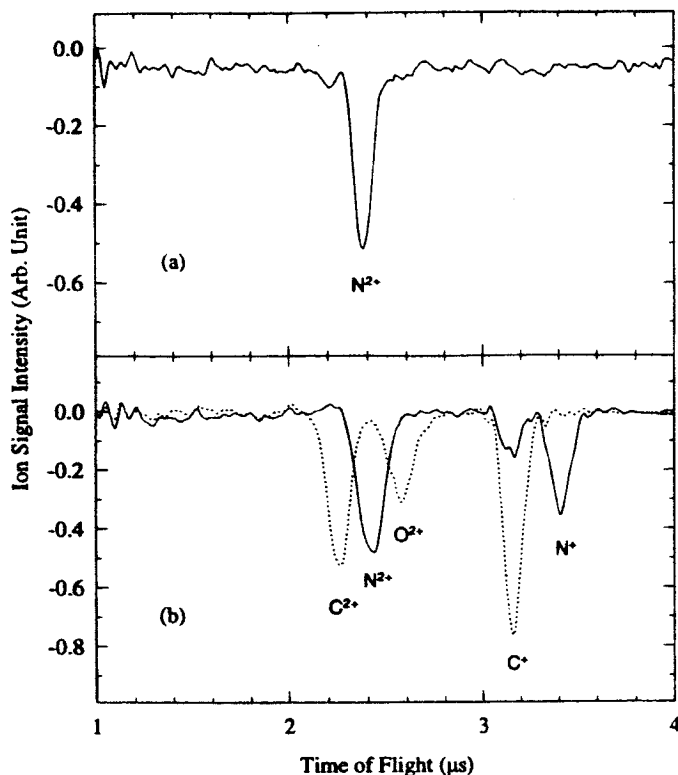


FIG. 1.—(a) TOF mass spectrum of the laser produced N_2^+ ions which have been stored in the trap. The parameters of the trap were set to exclude the ions with $m/q \geq 14$. (b) TOF mass spectra of the stored ions produced by electron impact dissociative ionization of N_2 (solid line) and CO (dotted line) gases. The parameters of the trap were set to store ions with their m/q up to 14.

to their extraction. Figure 1a is a typical TOF mass spectrum of the laser-produced ions stored in the trap under abovementioned trapping condition. Only N_2^+ ions are stored in the trap. For the purpose of comparison, Figure 1b shows the TOF mass spectra of stored N_2^+ , N^+ , C^{2+} , O^{2+} , and C^+ ions, produced by electron impact on ultrahigh-purity N_2 and CO gases. The number of N_2^+ ions stored in the trap was about 10^3 . The storage time ($1/e$) was in excess of 10 s at a base pressure of 4×10^{-10} torr.

The charge transfer rates were obtained by measuring the relative number of N_2^+ ions remaining in the trap as a function of time, in the presence of helium gas of known density. To minimize both the short-term and the long-term ion signal fluctuation and drift caused by the variation of laser power and the changing surface condition of the target when the ablation laser gradually drills into the target surface, the ion signals were measured, alternately, at a delay time t and at the shortest delay time $t_0 = 0.4$ s. Each pair of measurements consisted of 20 laser shots. More than 10 such pairs of measurements were made for each time t . The intensity ratio, $I_t = I_t/I_{t_0}$, is computed to obtain a normalized relative intensity. The storage time, t , is then scanned with a delay time increment, δt , to obtain an ion decay curve.

In this measurement, helium gas with ultrahigh-purity grade was used (Linde, ultrahigh purity, 99.999%). Helium gas was introduced into the vacuum chamber through a gas handling system. The operation of gas handling system to minimize contamination was described earlier (Fang & Kwong 1995). The purity of the He gas in the vacuum chamber was confirmed by a quadrupole mass analyzer.

3. DATA ANALYSIS AND RESULTS

Figure 2 is a plot of the relative ion signal intensities versus storage time, t , after laser ablation at four different helium gas pressures. The error bar for each data point represents the standard error (1σ), derived statistically from the measurements resulted by hundreds of laser shots. Each solid line is a weighted least-squares fit to the data by a single exponential decay function. The slope of the \ln -linear fit gives the charge transfer rate at a given helium pressure. The uncertainty of the charge transfer rate is determined by the weighted least-squares fit algorithm.

Figure 3 is a plot of the charge transfer rate between N_2^+ ion and He as a function of helium pressures. The slope in the figure is obtained by the weighted least-squares fits to a linear function. The charge transfer rate coefficient for N_2^+ ion and He obtained from slope of Figure 3 is $8.67(0.76) \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$. The uncertainty presented is the quadratic sum of the uncertainty of the helium density measurements (8%) and the uncertainty due to the statistical fluctuation of the ion intensities derived from the fitting.

4. DISCUSSION AND SUMMARY

4.1. Electronic State of the Stored N_2^+ Ions

N_2^+ ions produced by laser ablation can be in a variety of excited electronic states immediately after they are produced. They rapidly cascade to their ground state and low-lying metastable states through allowed transitions and collisional deexcitations by plasma electrons. We have shown in our previous publications that the ions produced by laser ablation at low laser power densities are in their ground state (Kwong & Fang 1993; Fang & Kwong 1994, 1995). Furthermore, N_2^+ has only one low-lying metastable state, $2s2p^2(^4P)$, with three fine-structure levels. Their lifetimes are: 0.98 ms for $^4P_{1/2}$, 13.4 ms for $^4P_{3/2}$, and 3.2 ms

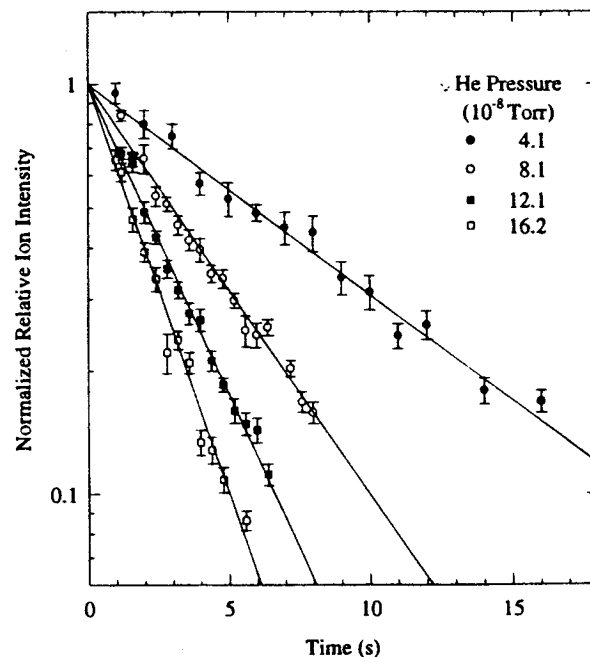


FIG. 2.—The decay curves of normalized relative intensity of N_2^+ ions vs. storage time at four different pressures of He. The uncertainty (1σ) is due to the statistical fluctuation of the ion signal. Solid lines are the least-squares fits to a single exponential function.

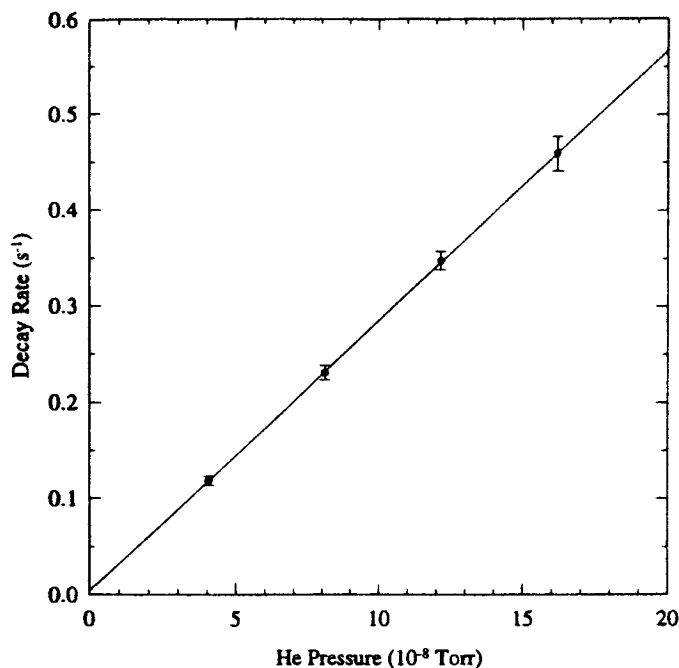


FIG. 3.— N^{2+} ion decay rate vs. He pressure. Each error bar represents the statistical uncertainty of 1σ . The slope of the straight line fit gives the charge transfer rate coefficient of the N^{2+} ion with He.

for $^4P_{5/2}$ (Fang, Kwong, & Parkinson 1993). At the time we start to measure the ion intensity (0.4 s after the laser ablation), the stored metastable N^{2+} ions, if there are any, should have decayed to the $2s^22p(^2P^o)$ ground state.

4.2. Mean Energy and Temperature

The mean energy of the stored N^{2+} ions is about 2.7 eV which is estimated from the potential well depth of the trap (Kwong & Fang 1993; Knight & Prior 1979). This mean energy corresponds to an ion temperature of about 2.1×10^4 K, calculated by using the relation $\bar{E} = 3/2kT$ since the velocity distribution of the ions in a trap is nearly thermal (Knight & Prior 1979). Considering the temperature of the He gas in the experiment is about 300 K (room temperature), it is more appropriate to introduce an equivalent temperature to compare with results where both reactants are in thermal equilibrium (Dickinson 1995). The equivalent temperature, T_{equiv} , is determined by the mean relative velocity between ion and neutral atom with the ion temperature T_i and the neutral temperature T_n . The relation

between these temperatures is given by

$$\frac{T_{equiv}}{\mu} = \frac{T_i}{m_i} + \frac{T_n}{m_n},$$

where m_i and m_n are the mass of ion and the mass of neutral, respectively, and μ is their reduced mass. The equivalent temperature for this measurement is 3.9×10^3 K.

4.3. Summary

Figure 4 illustrates the comparison of our measured charge transfer rate coefficient at temperature 3.9×10^3 K with the calculated total rate coefficient by Sun et al. (1996). The calculated value is within 1σ of our measurement.

We acknowledge the technical assistance of Heinz Knocke and Milton Lewis. This work is supported by FY 95 Nevada EPSCoR Laser Physics Program and NASA under contract NAGW-4279.

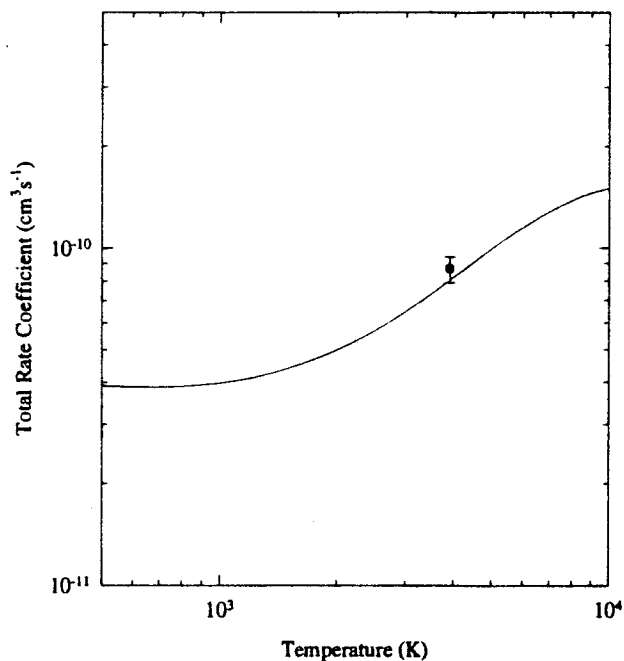


FIG. 4.—Total rate coefficients of charge transfer between N^{2+} ($2p^2P^o$) and He. The filled circle represents the present work. The solid line is the calculated result by Sun et al. (1996).

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Charge transfer between ground-state N^{2+} and H_2 , N_2 and CO at electron-volt energies

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(19 August 1996)

Abstract

The charge transfer rate coefficients for reactions of $N^{2+}(2s^22p^2P^o)$ with H_2 , N_2 , and CO are measured using ion storage. A cylindrical rf ion trap was used to store N^{2+} ions produced by laser ablation of a solid titanium nitride target. The rate coefficients were derived from the decay rate of the ion signal. The rate coefficients for the above three reactions are $3.38(0.35) \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$ at $T_{\text{equiv}} = 2.9 \times 10^3 \text{ K}$, $2.10(0.18) \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$ at $T_{\text{equiv}} = 1.3 \times 10^4 \text{ K}$, and $3.37(0.29) \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$ at $T_{\text{equiv}} = 1.3 \times 10^4 \text{ K}$, respectively. No theoretical or other experimental values are available at this energy range.

PACS numbers: 34.70.+e, 32.80.Pj, 52.50.Jm, 95.30.Dr

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I. INTRODUCTION

Charge transfer at electron-volt energies between multiply charged atomic ions and neutral atoms and molecules is of considerable importance in astrophysics, plasma physics and, in particular, fusion plasmas. This process can determine the ionization equilibrium in these plasmas. Despite its importance, however, very few experimental studies have been made at this energy range. Moreover, theoretical treatments on these low energy collisions are far from complete. Furthermore, only a few theoretical studies have been made on reactions with molecules [1,2]. This may be due to, in part, the complicated electronic, vibrational, and rotational structures of the molecules and the availability and the accuracy of the position and orientation sensitive potentials of the pseudo-molecule formed during the collision. Most of these calculations have not been tested. The experimental study of these processes is therefore particularly useful, not only to explain the ionization structures in these plasmas but also to verify the theoretical treatments at this energy range.

N^{2+} is of particular interest since it is present in significant abundance in astrophysical nebulae, nova shells, and interstellar molecular clouds [3,4]. These ions may play an important role in the production of excited state N^+ . Furthermore, molecules such as nitrogen may be present as impurities at the edge of the fusion reactors and inside the diverter of the future International Tokamak Experimental Reactor (ITER). Since N^{2+} is only 44 eV above its neutral ground state, it can easily be produced by a variety of mechanisms such as electron impact ionization, dielectronic recombination, photoionization, and charge transfer. Their electron-transfer collisions with helium, atomic and molecular hydrogen, nitrogen, carbon monoxide and other molecules at mean collision energies of a few electron-volts can play an interesting role in regulating the energy and therefore the characteristics of the plasmas in these regions.

Our laser ablation ion source and ion storage facilities are primarily designed to study these charge transfer reactions involving multiply charged atomic ions and a variety of neutral atoms and molecules at electron-volt energies. While our measured rate coefficient for O^{2+}

with helium [5,6] is orders of magnitude different from the full quantal calculation of Gargaud *et al.* [7] and Kimura *et al.* [8] and the Landau-Zener approximation of Butler *et al.* [9], our recent charge transfer measurement of N^{2+} with helium [10] is in good agreement with the calculation of Sun *et al.* [3]. These measurements may provide valuable information for the understanding of the collisional process and perhaps may inspire improvements in both experimental and theoretical approaches. In this paper, we will present charge transfer results of ground state ($2s^2 2p^2 P^o$) N^{2+} and H_2 , N_2 and CO at electron-volt energies.

II. EXPERIMENTAL METHOD

The techniques of the laser ablation ion source and ion storage have been described in detail in previous publications [6,10–12]. The N^{2+} ions were produced by laser ablation of solid titanium nitride (TiN) targets. We chose titanium nitride as the target material because of the distinct difference of the mass-to-charge ratio between N^{2+} ($m/q = 7$) and other low-charge state ions that could be produced simultaneously during laser ablation. These ions include N^+ ($m/q = 14$), Ti^+ ($m/q = 48$), Ti^{2+} ($m/q = 24$), and Ti^{3+} ($m/q = 16$), and will be the focus of other experiments in the future. We used approximately 1 mJ of the second harmonic (532 nm) output emission of a pulsed Nd:YAG laser as the ablation source. The power density of the laser at the ablation target surface was estimated to be about 10^8 W cm^{-2} . The use of low laser power density eliminated the formation of higher charge state ions of titanium and nitrogen.

The laser produced ions were selectively stored in an rf quadrupole trap by choosing the following trap parameters: rf $f = 1.44$ MHz, ac amplitude $V_0 = 350$ V, and dc bias $U_0 = 29$ V. The characteristics of the trap can be found in a previous publication [11]. These parameters generated a pseudo-potential well for N^{2+} with an axial well depth $D_z = 26.6$ eV and a radial well depth $D_r = 35.6$ eV. At these trap parameters, the low charge state of titanium ions and the singly-charged nitrogen ions were excluded from the trap.

The stored ions in the trap were analyzed by extracting all the ions from the trap by

two push-pull voltage pulses (+190 V and -200 V) applied simultaneously at the end caps of the trap. The extracted ions were then injected into a 0.3-m time-of-flight (TOF) mass spectrometer. They were collected and detected by a 1-in.-diameter microchannel plate. The TOF mass spectrum was recorded by a transient digitizer and stored for later analysis. While the time of flight of the ion signal was used to identify the type of ions, the amplitude of the ion signal was used to measure the N^{2+} population stored in the trap prior to their extraction. The TOF mass spectrum confirmed that only N^{2+} ions were stored in the trap. We estimated that the number of N^{2+} ions stored in the trap was about 10^3 . The storage time ($1/e$) was in excess of 10 s at the base pressure of 4×10^{-10} Torr.

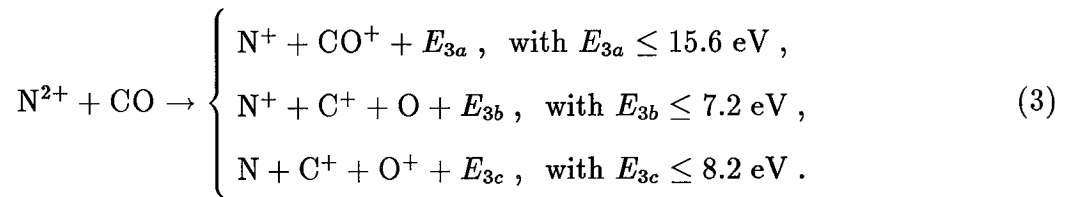
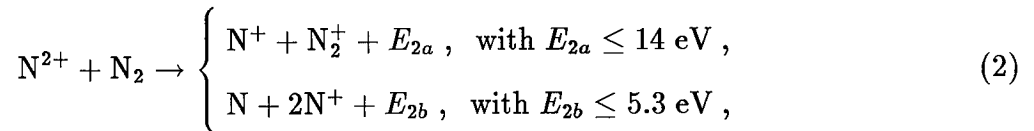
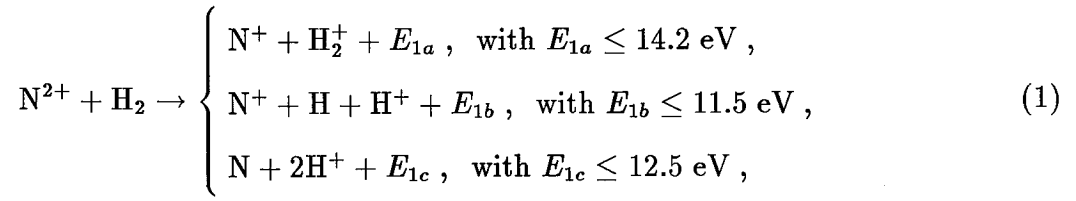
To measure the relative number of N^{2+} ions remaining in the trap, as a function of time after laser ablation, we measured the ion signal intensity alternately at a delay time t and at the shortest delay time $t_0 = 0.4$ s. More than a hundred such laser-ablation detection cycles for each delay time were carried out. The mean relative intensity, $I_r(t) = \frac{1}{N} \sum_{n=1}^N [I(t)/I(t_0)]_n$, and its statistical uncertainty were computed. This averaging procedure enables us to minimize the measurement error due to the short-term fluctuation and the long-term drift of the ion signal caused by the random change of the laser power and the gradual change of the surface conditions of the target as the ablation laser drills into the target. The storage time is then scanned at a later time t with a time increment δt . To obtain an ion decay curve, we measured the relative intensity of the ion signal at more than ten different storage times over one decade of the initial value at the shortest delay.

The charge transfer rate coefficients were obtained by measuring the decay rates of the stored ions in the presence of a reactant gas of known density (see Section III). Measurements obtained from four different gas pressures were usually adequate to derive accurately the rate coefficient. Since the laser ablation ion source uses a solid target in the ion production, the complexities and uncertainties caused by the interaction between the ions and their parent gas, as in some conventional ion source, are totally eliminated. In all the measurements, ultrahigh-purity (UHP) grade or research grade reactant gases were used. These gases include H_2 (Alphagaz, UHP, 99.999%), N_2 (Airco, UHP, 99.999%), and CO (Alphagaz,

research, 99.97%). The reactant gas was introduced into the vacuum chamber through a stainless steel gas handling system. The procedure for the operation of the gas handling system to minimize contamination was described earlier [6]. The purity of the neutral reactant gas in the reaction chamber was further confirmed by a Vacuum Generator DX100 quadrupole residual gas analyzer.

III. DATA ANALYSIS AND RESULTS

The ground state of N^{2+} is 44 eV above its neutral ground state. The reaction channels with both single and double electron capture from reactant molecular gas are energetically possible, for example:



However, the parameters of the ion trap were set only to store N^{2+} . Ions with m/q greater than 10 or smaller than 4 are outside the stable region of the trap and will not be stored. Low charge state titanium ions, N^+ ions formed during laser ablation, and all product ions produced by single electron transfer and double electron transfer were also excluded from the trap. For this measurement, our present facility only allows us to measure the total charge transfer rate coefficient. Improvement on the facility to identify the energy state of the product ions is underway.

The N^{2+} ions were stored in a 27 eV potential well, the probability of N^{2+} being kicked out of the trap by elastic collisions with reactant gas at room temperature (0.04 eV) is very small. This loss mechanism could be ignored in the analysis.

The time dependent mean relative intensity, $I_r(t)$, of the stored ions is related to the total charge transfer rate coefficient K by a single-exponential function:

$$I_r(t) = \exp[-R(t - t_0)] , \quad (4)$$

with

$$\begin{aligned} R &= n_m \langle v_1 q_1 \rangle + n_b \langle v_2 q_2 \rangle \\ &= n_m K + R_b , \end{aligned} \quad (5)$$

where n_m is the density of the reactant gas, n_b is the density of the residual background gas in the ultra-high vacuum chamber, $I_r(t)$ is proportional to the number of N^{2+} ions at time t , q_1 and q_2 are the charge transfer cross sections to all channels including single and multiple electron transfer with the reactant molecular gas and with residual gas respectively, and v_1 and v_2 are the relative velocities of the interacting multiply charged ions with reactant gas molecules and the residual gas molecules respectively. R_b is the decay rate of N^{2+} in the ultra-high vacuum system with only residual background gas present. The data set containing the mean relative intensity $I_r(t)$ (cf. Sec. II) was fitted to Eq. (4) using the weighted least-squares method. The weight for each point is determined from its statistical uncertainty. The decay rate R , as a parameter of the fitting, and its statistical uncertainty, were determined from the fitting algorithm. The rate coefficient K was obtained by the weighted least-squares fits of the decay rates R at each reactant molecular gas density n_m to Eq. (5). The uncertainty of the rate coefficient was a quadratic sum of the statistical uncertainty derived from the data fitting and the uncertainty of the reactant gas density measurement. Absolute calibration of the ion gauge to measure the reactant gas density has been described in a previous publication [11] and will not be elaborated here.

The data and their fitting curves for the reaction $N^{2+} + H_2 \rightarrow$ products are shown in Figs. 1(a) and 1(b). Figure 1(a) shows the decay of the relative N^{2+} intensities at four different

H₂ pressures. The solid lines represent the result of the weighted least-squares fit. Figure 1(b) plots the decay rates as a function of H₂ pressure. The slope of the solid line gives the charge transfer rate coefficient of $3.38(0.35) \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$. In a similar way, Figures 2(a) and 2(b) show the results of the reaction $\text{N}^{2+} + \text{CO} \rightarrow \text{products}$. The rate coefficients of N^{2+} with CO is $3.37(0.29) \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$.

The N^{2+} with N_2 measurement was carried out with two different ion sources: (1) laser ablation ion source as was described in Sec. II and (b) the electron impact ionization ion source with N_2 as the parent gas. The results of these two measurements are consistent: $2.10(0.18) \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$ for the laser ablation method, and $2.0(0.2) \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$ for the electron impact ionization method. These results are also consistent with our previous result of $1.73(0.18) \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$ [11]. Both measured results are in agreement with that obtained by Church and Holzscheiter [13]. Figures 3(a) and 3(b) show the results using the laser ablation method.

IV. DISCUSSION

The N^{2+} ions produced by laser ablation can be in a variety of excited electronic states immediately after they are produced. They rapidly cascade to their ground state and low lying metastable states through allowed transitions and collisional deexcitations by plasma electrons. We have shown in previous measurements that the ions stored in the trap produced by low laser power are in their ground state [5,6,12]. Furthermore, N^{2+} has only one low lying $2s2p^2(^4P)$ metastable state with three fine structure levels. Their lifetimes are: 0.98 ms, 13.4 ms and 3.2 ms for the 4P_j with $j = \frac{1}{2}, \frac{3}{2}, \text{ and } \frac{5}{2}$ respectively [14]. At the time we start to measure the ion intensity (0.4 s after the laser ablation), the stored metastable N^{2+} ions, if there are any, should have decayed to the $2s^22p(^2P^o)$ ground state.

The mean energy of the stored N^{2+} ions is about 2.7 eV [5,15]. This mean energy corresponds to an ion temperature of about $2.1 \times 10^4 \text{ K}$, which can be calculated by using the relation that the mean energy \bar{E} and the temperature T is related by $\bar{E} = \frac{3}{2}kT$ since the

velocity distribution of the ions in an rf trap is nearly thermal [15]. Because the temperature of the neutral reactant gas is at room temperature (300 K), we introduce an equivalent temperature corresponding to the mean relative velocity of the N^{2+} and the reactant gas molecules [16]. This equivalent temperature T_{equiv} of the collisional system is given by

$$\frac{T_{\text{equiv}}}{\mu} = \frac{T_i}{m_i} + \frac{T_n}{m_n},$$

where the ion temperature is T_i and the reactant gas temperature is T_n . m_i and m_n are the masses of the ion and the neutral respectively, and μ is their reduced mass. Using this relation, the equivalent temperatures for each measurement are estimated to be 2.9×10^3 K for N^{2+} with H_2 , 1.3×10^4 K for N^{2+} with CO and N_2 .

The above data show the charge transfer reaction for N^{2+} with H_2 is slower by two orders of magnitude than that for N^{2+} with CO and N_2 , even though all three reactant molecules have similar ionization potentials. Furthermore, the charge transfer reaction with H_2 is slower than that with He [10]. This could presumably be due to the sparsity of low-lying electronic states of H_2^+ .

ACKNOWLEDGMENTS

We acknowledge the technical assistance of Heinz Knocke. Helpful discussions with Phillip Stancil are appreciated. This work is supported by FY 95 Nevada EPSCoR Laser Physics Program and NASA under contract NAGW-4279.

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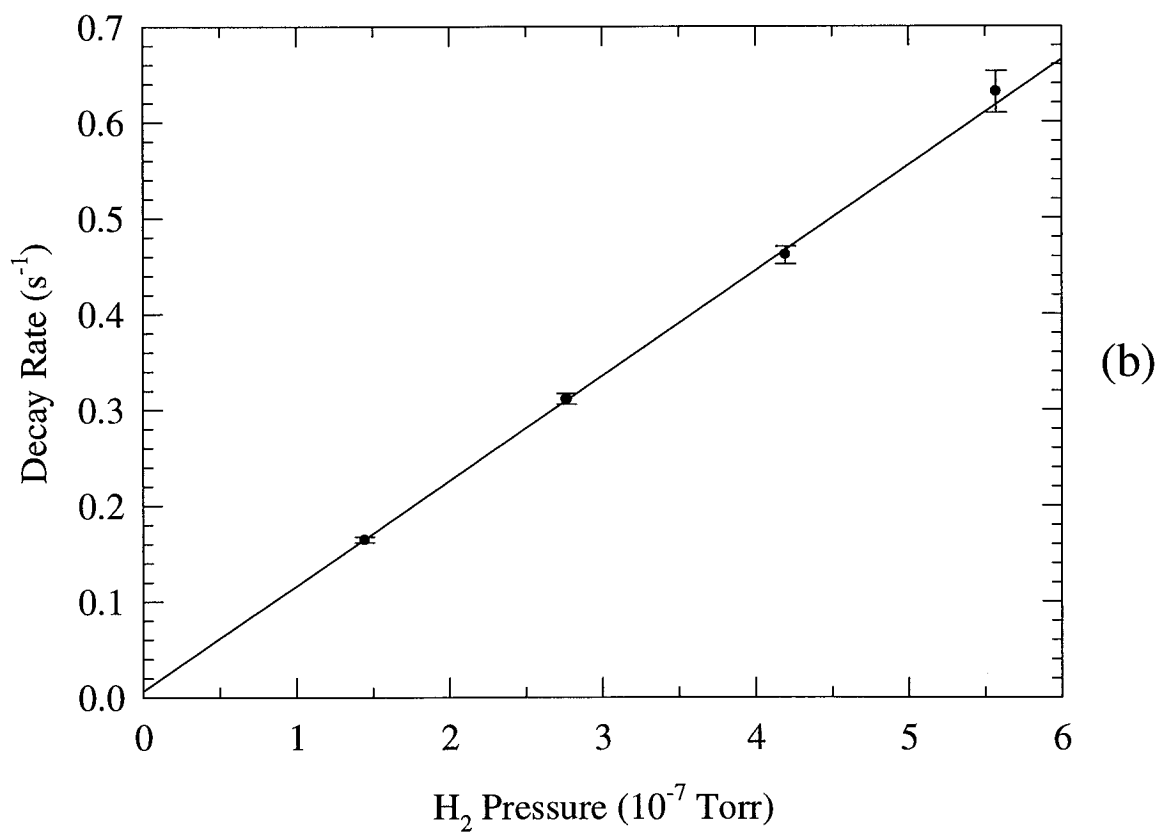
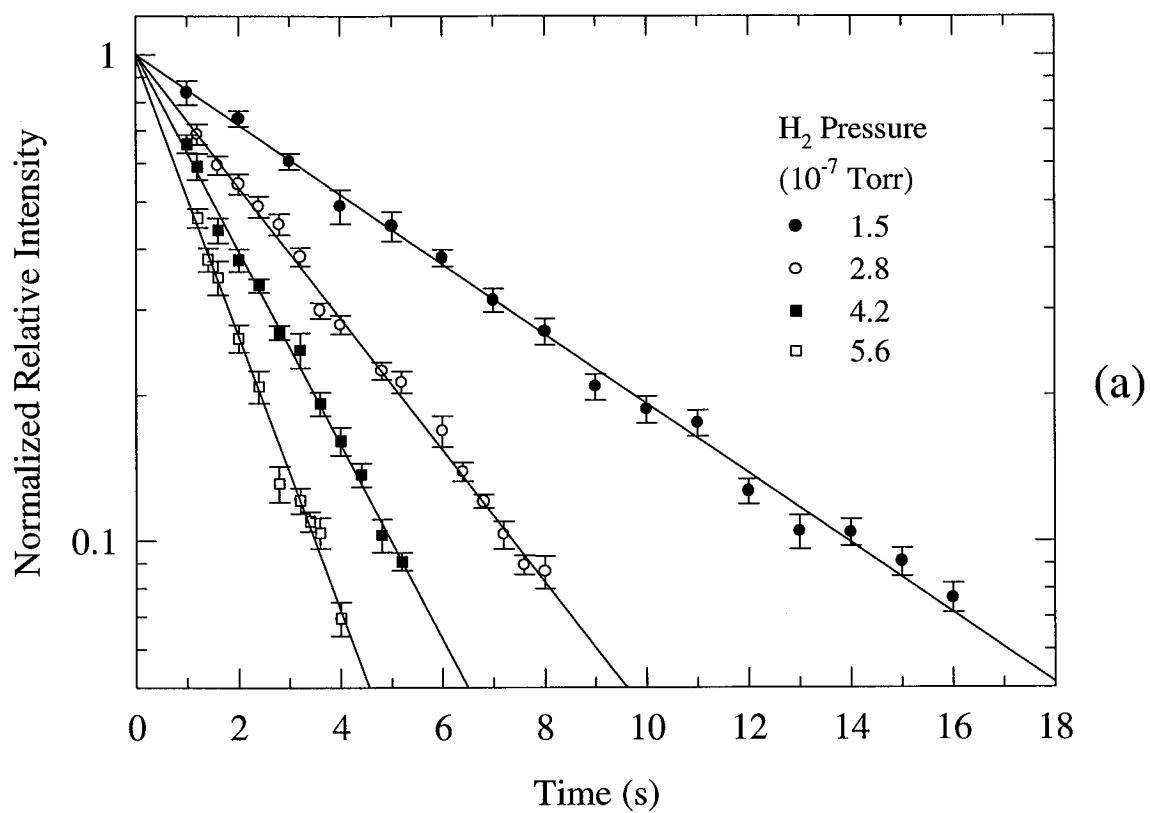
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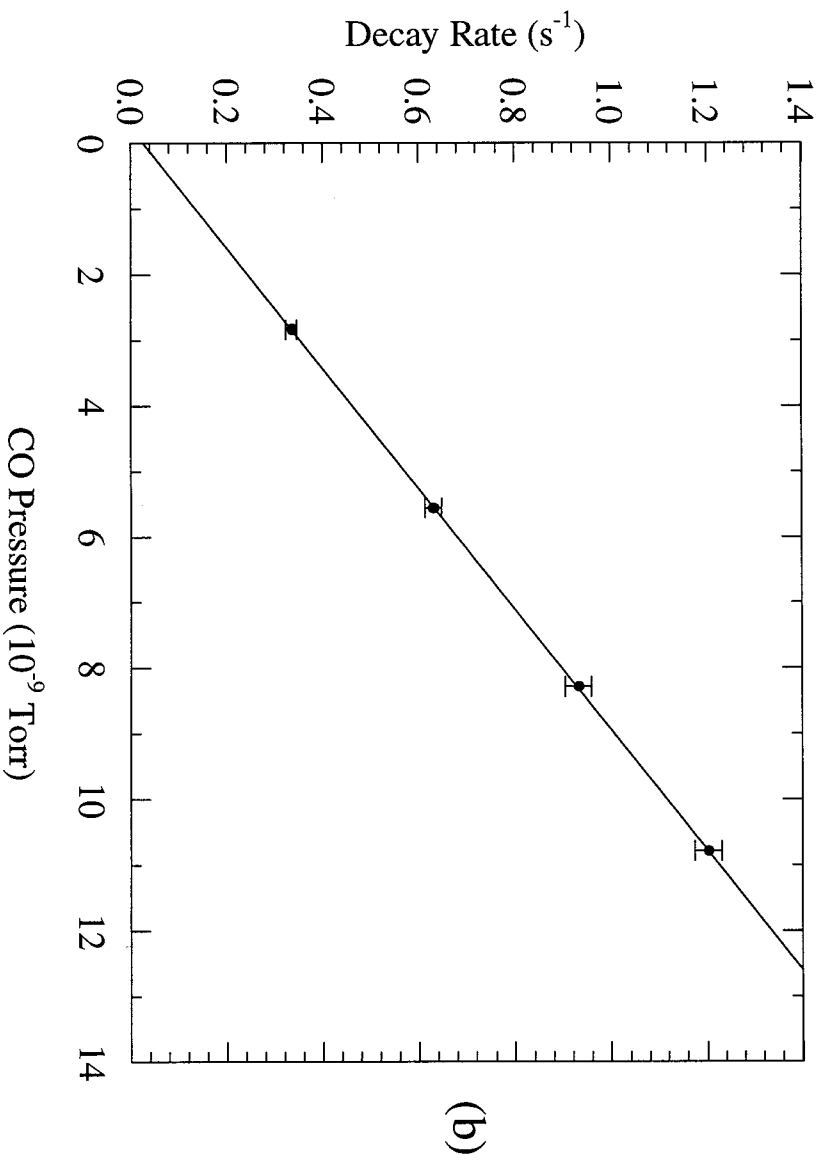
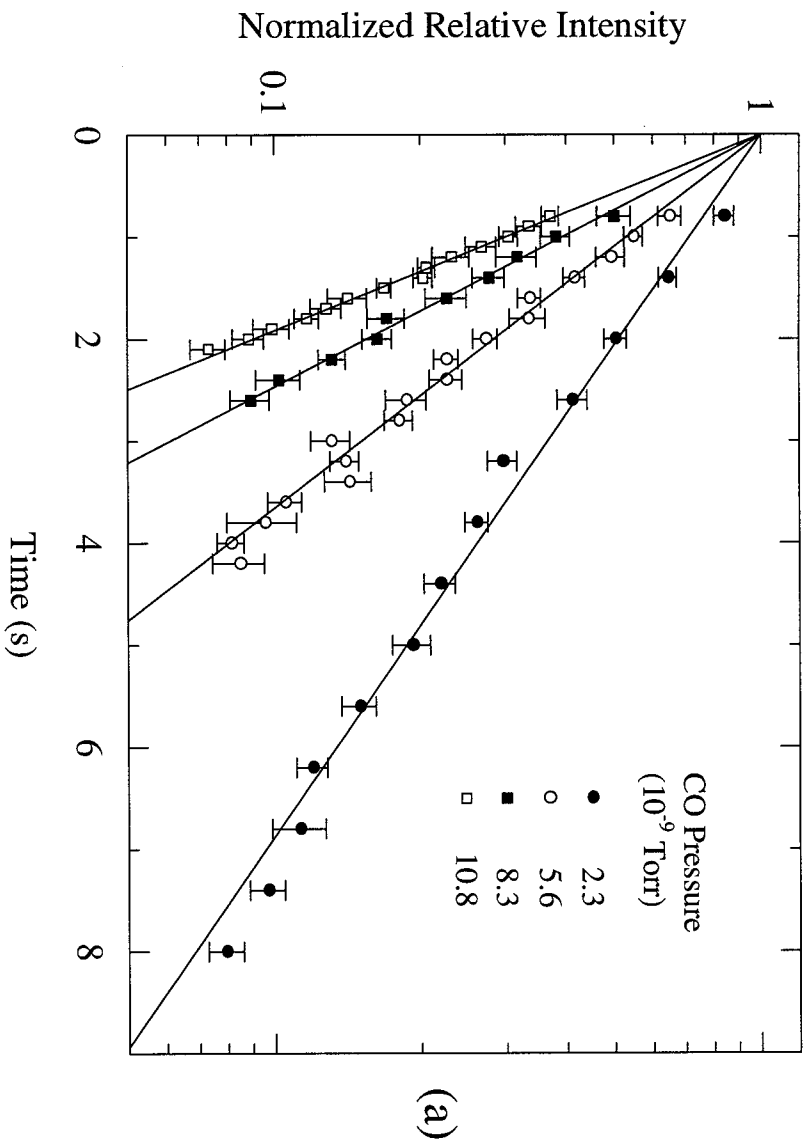
FIGURES

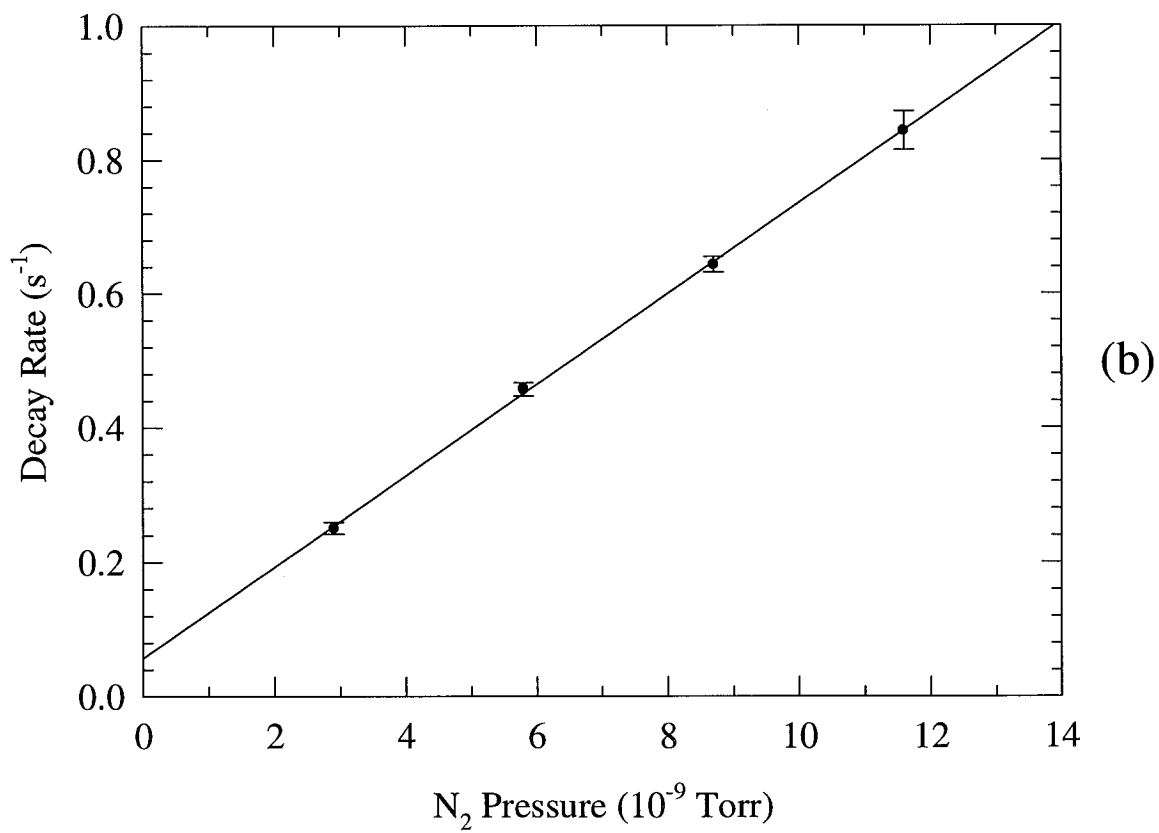
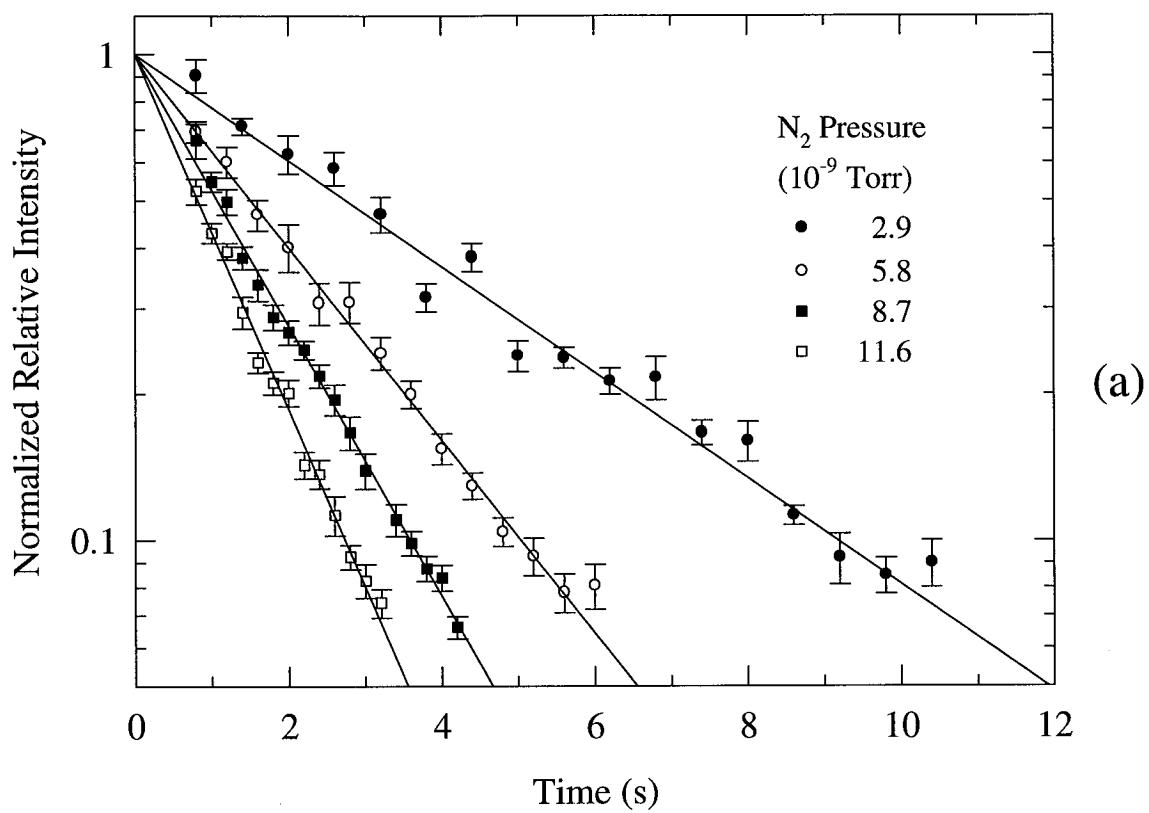
FIG. 1. (a) The normalized relative intensity of N^{2+} vs storage time at four different pressures of H_2 . Solid lines are the least-squares fits to a single exponential function. (b) N^{2+} decay rate vs H_2 pressure.

FIG. 2. (a) The normalized relative intensity of N^{2+} vs storage time at four different pressures of CO. (b) N^{2+} decay rate vs CO pressure.

FIG. 3. (a) The normalized relative intensity of N^{2+} vs storage time at four different pressures of N_2 . (b) N^{2+} decay rate vs N_2 pressure.







Charge transfer between the $2p^2\ ^3P$ ground-state O^{2+} ion and He, H_2 , N_2 , and CO at electron-volt energies

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The charge-transfer rate coefficients for the reaction $O^{2+}(2p^2\ ^3P) + He, H_2, N_2, \text{ and } CO \rightarrow$ products are measured at energies about 2.5 eV using a laser-induced-plasma ion source and ion storage. The rate coefficients are $1.12(0.15) \times 10^{-12} \text{ cm}^3 \text{ s}^{-1}$, $2.36(0.22) \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$, $3.15(0.26) \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$, and $3.40(0.29) \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$, respectively. While our result for O^{2+} and H_2 is consistent with the published value, the charge-transfer rate coefficient for O^{2+} and He disagrees with all the available theoretical and experimental values by as much as three orders of magnitude. No theoretical value is available for $O^{2+} + N_2$. However, our measured value at 2×10^4 K is different from that obtained by the drift tube method at 300 K by a factor of 2. No theoretical and experimental values are available for comparison for the charge-transfer reaction of O^{2+} and CO.

PACS number(s): 34.70.+e, 32.80.Pj, 52.50.Jm, 95.30.Dr

I. INTRODUCTION

Charge-transfer processes between multiply charged ions and neutral atoms and molecules are of considerable interest in astrophysics, plasma physics, and for verifying basic theoretical treatments in low energy collisions. The O^{2+} ions are of particular interest since they are abundant in the terrestrial ionosphere, in supernovas, and in the upper atmospheres of Mars and Venus [1], and in the upper atmospheres of Earth [2]. Their electron-transfer collisions with He, H_2 , and N_2 , at mean collision energies of a few eV or less, often play important roles in the formation and destruction of ions related to them. However, our recent charge-transfer measurement of O^{2+} and He [3] differs significantly from the calculations of Dalgarno *et al.* [1] and Gargaud *et al.* [4]. In this paper, we will re-examine our results, and will provide additional evidence to support the result of our previous measurements of the $2p^2\ ^3P$ ground-state O^{2+} ion and He. Furthermore, we will present charge-transfer results of the $O^{2+}(2p^2\ ^3P)$ ion and H_2 , N_2 , and CO at electron-volt energies using laser ablation and the ion trap storage technique.

II. EXPERIMENTAL METHOD

The measurements of the ground-state thermal energy charge-transfer rate coefficient between O^{2+} ions and molecules are carried out by using a technique which combines a laser ablation ion source and ion storage. With this technique, ions are produced from a solid target, and no source gas is required. This eliminates the potential source of systematic error due to reactions between ions and the source gas. 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 experiment have been de-

scribed in detail earlier [5,6]. Briefly, O^{2+} ions are produced by laser ablation of solid ferric oxide (Fe_2O_3) targets. We used the output of the second harmonic (532 nm) of a pulsed Nd:YAG laser (Molelectron MY-34) as an ablation source. In this series of measurements, approximately 0.3 mJ of the second harmonic of the Nd:YAG laser was used. The power density was estimated to be about 10 MW cm^{-2} . The low power density of the ablation source eliminates the formation of higher charge states of iron and oxygen, which can be stored simultaneously with O^{2+} ions in the trap. The trapping parameters (rf frequency $f = 1.4 \text{ MHz}$, amplitude $V_0 = 380 \text{ V}$, and dc bias $U_0 = 35 \text{ V}$) of the rf quadrupole ion trap were chosen to selectively store O^{2+} ions. Lower charge states of iron and oxygen ions were excluded from the trap. The number of O^{2+} ions stored in the trap was about 10^3 , and the mean energy of the ion was estimated to be about 2.5 eV ($2 \times 10^4 \text{ K}$) [3,7]. The storage time ($1/e$) was in excess of 10 s at a base vacuum pressure of $5 \times 10^{-10} \text{ Torr}$. The charge-transfer rate measurements were performed by measuring the relative number of O^{2+} ions remaining in the trap, as a function of time, after they were stored in the presence of a neutral gas of known density. The O^{2+} ions were extracted from the trap and their relative number was measured by a 0.3-m time-of-flight (TOF) mass spectrometer equipped with an electrostatic lens and a 1-in.-diameter dual microchannel plate.

In all the measurements, neutral gases with a minimum of ultrahigh-purity grade were used. These gases include He (Linde, ultrahigh purity, 99.999%), H_2 (Alphagaz, ultrahigh purity, 99.999%), N_2 (Airco, ultrahigh purity, 99.999%), and CO (Alphagaz, research, 99.97%). The neutral gas was introduced into the vacuum chamber through a gas handling system. To minimize contamination during the transfer of the gas into the vacuum chamber, the reservoir of the gas handling system was evacuated to $\sim 10^{-2} \text{ Torr}$ through a cold trap with a mechanical pump. The reservoir was then flushed with

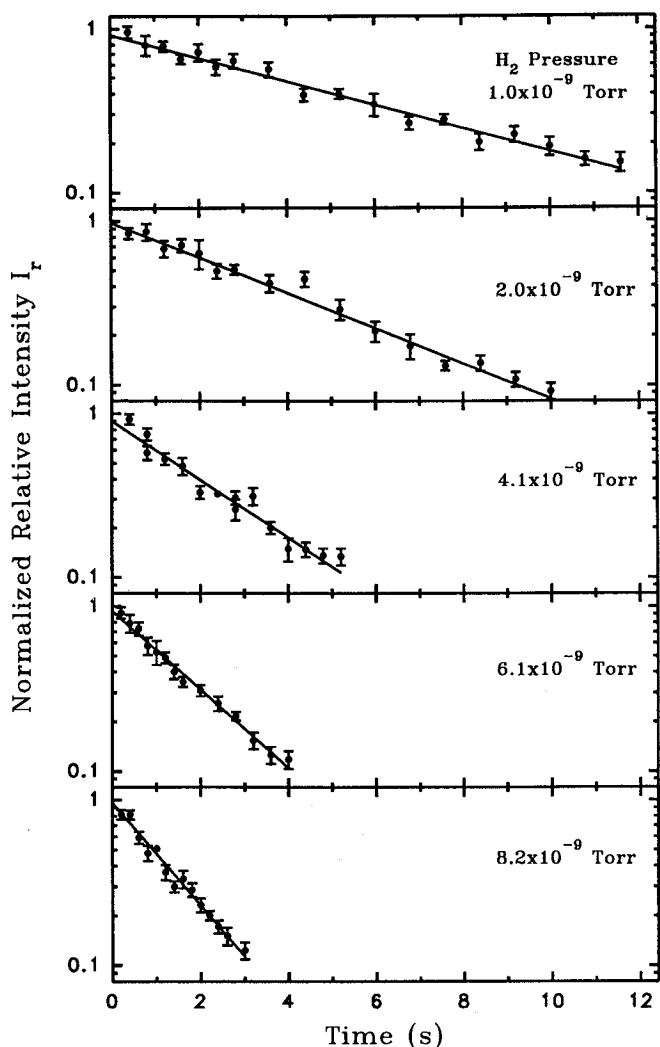


FIG. 2. The decay curves of normalized relative intensity of O^{2+} ion vs storage time at five different pressures of H_2 . Each data point represents about 130 measurements. The uncertainty (1σ) is due to the statistical fluctuation of the ion signal. Solid lines are the least-squares fits to a single exponential function.

If the charge-transfer rate coefficient for the ground-state O^{2+} ion and He is as high as $10^{-9} \text{ cm}^3 \text{ s}^{-1}$, then even at the lowest He pressure of 0.5×10^{-6} Torr used in our recent measurement [3], nearly all the ground-state O^{2+} ions would have captured electrons and would have left the trap within 0.4 s after the O^{2+} ions were created and stored in the trap. Those O^{2+} ions that were left behind in the trap and reacted slowly with He should have been in their low lying metastable states. The only metastable state that can survive for 0.4 s or more in the trap must be the long-lived $2p^2\ ^1D$ metastable state of O^{2+} ions. This argument, however, contradicts the result of our previous experiment with H_2 ; i.e., all O^{2+} ions are in their $2p^2\ ^3P$ ground state [6].

We have performed further tests to determine if the rapid charge-transfer rate of $10^{-9} \text{ cm}^3 \text{ s}^{-1}$ exists for the $O^{2+} + \text{He}$ reaction. If we assume such a rate coefficient, then at a He pressure of 5×10^{-8} Torr the mean charge-transfer collision time is about 0.6 s. We should be able

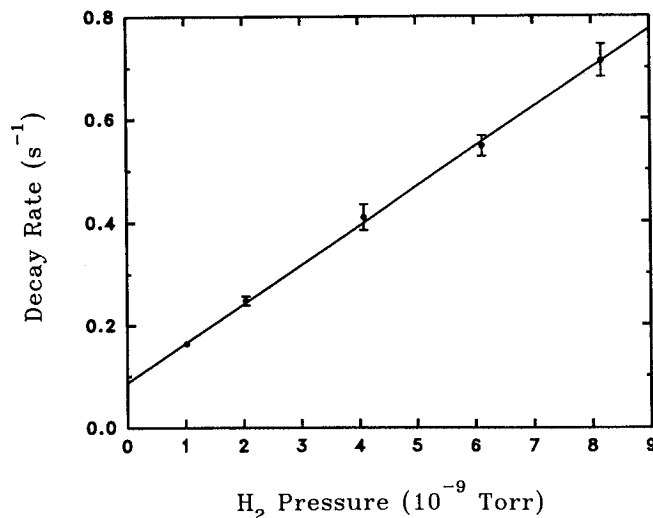


FIG. 3. O^{2+} ion decay rate vs H_2 pressure. Each error bar represents the statistical uncertainty of 1σ . The slope of the straight line fit gives the charge-transfer rate coefficient of the O^{2+} ion with H_2 .

to detect a significant drop in the ion signal due to such a fast reaction rate. We have eliminated the change in the ion signal caused by the residual gases in the vacuum chamber by comparing the O^{2+} ion signal intensity in the presence of 5×10^{-8} Torr of He and with He pumped out of the system. Figure 4 shows the ratio of O^{2+} ion signal intensities with 5×10^{-8} Torr of He and without He at a delay time of 0.4 s, 1 s, and 2 s after the ions are stored in the trap. The invariance of the differential ion signals over two seconds indicates that a rapid charge-transfer rate with He does not exist.

Since the charge-transfer rate between the ground-state O^{2+} ion and helium may be several orders of magnitude smaller than that of molecular gases, the purity

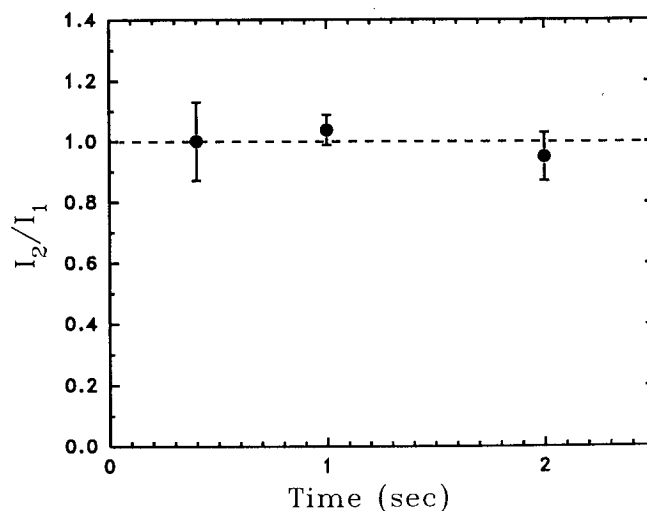


FIG. 4. The ratio of the O^{2+} ion signal, I_2 , at 5×10^{-8} Torr of He and the O^{2+} ion signal, I_1 , with He pumped out of the vacuum chamber at 0.4 s, 1 s, and 2 s after O^{2+} ions are stored in the trap. The horizontal dotted line indicates the intensity ratio of 1.

TABLE I. Measured and calculated charge-transfer rate coefficients K for O^{2+} with He, H_2 , N_2 , and CO.

| Reaction | Reference | K ($cm^3 s^{-1}$) | Temperature (K) | Method |
|----------------|------------------------------|---|------------------------------|-----------------------------|
| $O^{2+} + He$ | [12] | $\leq 10^{-14}$ | 3×10^2 | Experiment, drift tube |
| | [13] | $3.5(1.5) \times 10^{-11}$ | 4×10^2 | Experiment, drift tube |
| | [14] | 2×10^{-11} | 4×10^2 | Theory, distorted wave |
| | [1] | 10^{-14} | 3×10^2 | Theory, quantal calculation |
| | | 4×10^{-14} – 4×10^{-13} | 1×10^3 | |
| | | 2×10^{-11} – 1×10^{-10} | 1×10^4 | |
| | [15] | 8×10^{-12} | 3×10^2 | Theory, quantal calculation |
| | | 1.0×10^{-10} | 5×10^3 | |
| | | 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 | |
| | [4] | 0.7×10^{-10} | 2×10^2 | Theory, quantal calculation |
| | 1.3×10^{-10} | 5×10^2 | | |
| | 1.01×10^{-9} | 2×10^4 | | |
| [3] | $1.12(0.15) \times 10^{-12}$ | 2×10^4 ^a | Experiment, ion trap | |
| $O^{2+} + H_2$ | [9] | $1.71(0.15) \times 10^{-9}$ | 1×10^4 ^b | Experiment, Penning trap |
| | [6] | $2.36(0.22) \times 10^{-9}$ | 2×10^4 ^a | Experimental, Paul trap |
| $O^{2+} + N_2$ | [11] | $1.3(0.3) \times 10^{-9}$ | 3×10^2 | Experimental, drift tube |
| | This work | $3.15(0.26) \times 10^{-9}$ | 2×10^4 ^a | Experimental, Paul trap |
| $O^{2+} + CO$ | This work | $3.40(0.29) \times 10^{-9}$ | 2×10^4 ^a | Experimental, Paul trap |

^aEstimated from $\bar{E} = \frac{3}{2}kT$ with $\bar{E} \approx 2.5$ eV. Previous measurements in a rf trap indicate that the ion energy distribution is approximately thermal [7].

^bAlso estimated from the mean energy, which is $\bar{E} = 1.25$ eV, given in Ref. [9].

The rate coefficient for ground-state O^{2+} ions and He is $1.12(0.15) \times 10^{-12} cm^3 s^{-1}$.

V. MEASUREMENT OF O^{2+} IONS AND N_2 AND CO

Measurements were also carried out for the charge transfer between ground-state O^{2+} ions and N_2 and CO. In both cases, ultrapure grade N_2 and CO gases were used. The purity of these gases was confirmed using the same procedure described above. The charge-transfer rate coefficient (K) is derived using the same procedure as before; i.e., from the slope of the ion decay rate ($1/T$) vs gas density (P):

$$\frac{1}{T} = \frac{1}{T_0} + KP, \quad (1)$$

where $\frac{1}{T_0}$ is the charge-transfer rate coefficient of O^{2+} ions and the residual gas in the vacuum chamber ($\sim 5 \times 10^{-10}$ Torr). Figures 5 and 6 show the O^{2+} ion intensities as a function of storage time for N_2 and CO, respectively, at different gas pressures. The solid lines represent the least-squares fits of the data to a one-exponential function. The monotonic decrease in the collisional rate with decreasing target gas pressure is shown in Figs. 7 and 8. The slopes in the figures are obtained by a weighted least-squares fit to a linear function according to Eq. (1). The rate coefficients for ground-state O^{2+} ions and N_2 and CO are $3.15(0.26) \times 10^{-9} cm^3 s^{-1}$ and $3.40(0.29) \times 10^{-9} cm^3 s^{-1}$, respectively. The esti-

mated uncertainty of the result is mainly due to the uncertainty in the statistical fluctuation of the ion signals (typically about $\pm 2.5\%$) and the uncertainty in the estimation of the target gas density ($\pm 8\%$) [5]. While there are no available data for comparison with our CO measurement, our value for N_2 differs by a factor of 2 from results obtained using a drift tube at lower energies [11].

VI. SUMMARY AND RESULTS

Table I summarizes the results of various calculations and measurements. While our results for O^{2+} and H_2 are consistent with published values, our measured value for O^{2+} ions and He disagrees by as much as three orders of magnitude with the most recent calculation. Neither theoretical nor experimental values for CO are available for comparison. We hope that these measurements will stimulate further refinement of theoretical treatments, and improve the accuracy of charge-transfer measurements at electron-volt energies [16].

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