Charge transfer between C^{2+} and H$_2$, N$_2$, He, and CO at electron-volt energies

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We measured the total charge-transfer rate coefficients for C^{2+} with H$_2$, N$_2$, and CO using a cylindrical radiofrequency ion trap. C^{2+} ions were produced by the electron impact of residual CO in an ultrahigh-vacuum chamber. The rate coefficients for C^{2+} with H$_2$, N$_2$, and CO are measured to be 8.77 (0.71) $\times 10^{-11}$ cm^3 s^{-1} ($T_{\text{equiv}} = 2.63 \times 10^3$ K), 1.08 (0.07)$\times 10^{-10}$ cm^3 s^{-1} ($T_{\text{equiv}} = 1.17 \times 10^4$ K), and 4.58 (0.29) $\times 10^{-10}$ cm^{3}s^{-1} ($T_{\text{equiv}} = 1.17 \times 10^4$ K). The rate coefficient for C^{2+} with He is estimated to be less than 10^{-12} cm^{3}s^{-1} ($T_{\text{equiv}} = 4.38 \times 10^3$ K).

I. INTRODUCTION

In the interstellar cloud, multiply charged ions are formed and coexist with neutral atoms and molecules. Under astrophysical conditions that the interstellar gas is not in thermodynamic equilibrium because of its low density [1,2], ion-neutral charge-transfer reaction may play a dominant role in the distribution of elements among its various ionization stages [3–6]. Therefore, it is important to know the charge-transfer rate coefficients at the temperatures of these environments for modeling these astrophysical plasmas.

Carbon ions have been observed in a wide range of astrophysical regions and in fusion plasmas. Charge-transfer reactions of C^{2+} have been of interest for the past 20 years since it is typical of the many multiply charged species in these environments. In addition, a standing problem exists, i.e., an underabundance of C^{2+} and C$^+$ ions was observed in the HI region towards several unreddened stars (αEri, λSco, and νSco) [7], because the ratios of C^{2+}:C$^+$ and C$^+$:C in this region were observed to be inconsistent with radiative recombinations in a hot, partially ionized intercloud medium [7,8]. Charge-transfer reaction has been proposed [8] to explain the observed underabundance of C^{2+} and overabundance of C. In the case that this disparity is due to charge transfer between carbon ions and H atom, the charge-transfer rate coefficient for C^{2+} and C$^+$ has to be of the order of 10^{-11} cm^3/s and 10^{-12} cm^3/s, respectively. However, Mc-Carroll and Valiron [9] showed that charge transfer is unlikely to be important in the conversion of C^{2+} to C$^+$ and C$^+$ to C, because such charge-transfer probability estimated by the Laudau-Zener approximation is negligibly small at thermal energies. A quantum-mechanical calculation [10] agreed with McCarroll and Valiron [9] that the charge-transfer process of C^{2+} with H is slow with a rate coefficient of 1×10^{-12} cm^3/s. Recent *ab initio* calculations [11] for C^{2+} with H above 10 eV agree with that of [10] to the same order magnitude. In addition, the contribution of C$^+$ by the charge-transfer reaction between C^{2+} and He was excluded due to the slow reaction rate [2]. This, however, has not been confirmed experimentally.

In this paper, we report the first measurements to our knowledge of charge-transfer rate coefficients of ground state C^{2+} with H$_2$ and He, and ground state C^{2+} with N$_2$ and CO at electron-volt energies. The former two reactions may shed light on the problem of C^{2+} underabundance in the HI region. All these reactions with molecular gases and He can be important in astrophysical regions where carbon ions and these molecules coexist.

Charge-transfer reactions between C^{2+} and H$_2$, N$_2$, O$_2$, and CO at keV collisional energies have been investigated experimentally by Phaneuf *et al.* [12], Nutt *et al.* [13], Leputsch *et al.* [14], Burns *et al.* [15], and Unterreiter *et al.* [16]. To the best of our knowledge, no theoretical work and measurements have been reported for C^{2+} with H$_2$, N$_2$, He, and CO at electron-volt energies.

II. EXPERIMENTAL METHOD

The general experimental technique, which combines the ion trap and laser-plasma/electron beam ion source developed by Kwong *et al.* [17], has been described in detail in previous publications [18,19]. Since the facility has been upgraded over time and new procedures have been adopted for the measurement, procedures involving the current measurement can be found in the preceding article by Chen, Gao, and Kwong [20]. Here, only the specific use of the facility directly related to current measurements will be discussed.

In this measurement, C^{2+} ions were produced by the electron impact ionization of residual CO gas (4×10^{-11} Torr) in an ultrahigh-vacuum chamber. We did not use laser ablation of a carbon target to generate C^{2+} ions in this measurement because of the instability of the ion production. We were able to produce a measurable amount of C^{2+} ions from the residual CO in the vacuum chamber with an electron beam. The presence of a trace amount of CO in the reaction chamber does not introduce a measurable systematic error in our measurement. The unfocused electron beam used for ionization was created from a BaO dispenser cathode and was biased at a potential between -41 V and -150 V. The base pressure in the vacuum chamber was about 4×10^{-11} Torr with H$_2$ and CO as the major components. The mass resolution ($m/\Delta m$) of the TOF is about 23 at 6 amu.

The electron beam produced C^{2+} ions were stored in a cylindrical radiofrequency (rf) ion trap where charge transfer with the neutral atom/molecule of interest occurred. The trapping parameters (radiofrequency $f = 1.44$ MHz, amplitude $V_0 = 275$ V, and dc bias $U_0 = 20$ V) were chosen to se-
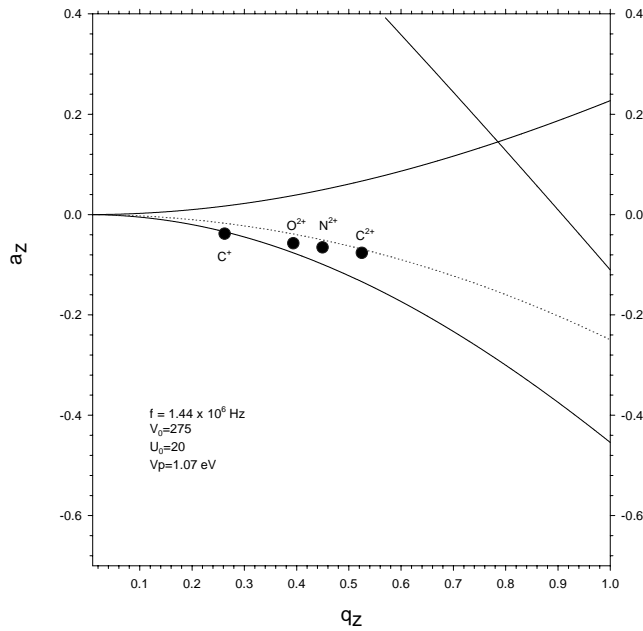
FIG. 1. The operating points of several ions in the stability diagram of the rf trap. Here a_z and q_z are the Mathieu parameters for the rf trap. The trapping parameters are $f = 1.44$ MHz, $V_0 = 275$ V, $U_0 = 20$ V, $qD_z = 10.73$ eV, and $qD_r = 12.68$ eV.

lectively store C^{2+} ions. The axial well depth was $qD_z = 10.73$ V, and the radial well depth was $qD_r = 12.68$ V. Since the difference in the mass-to-charge ratio between C^{2+} ($m/q = 6$) and N^{2+} ($m/q = 7$) is 1 amu, N^{2+} ions were also stored in the trap when N_2 was used as a target gas. Other ions produced by electron impact of the residual gases and target gases, i.e., O^{2+} ($m/q = 8$), C^+ ($m/q = 12$), O^+ ($m/q = 16$), and CO^+ and N_2^+ ($m/q = 28$), were almost excluded from the trap by a judicious choice of the trapping parameters, and the energy and duration of the electron beam. Figure 1 shows the typical operating points of C^{2+}, N^{2+}, C^+, and O^{2+} in the stability diagram of the rf trap [17]. While the operating point for C^{2+} is well within the stable region of the trap, the operating points for O^{2+}, C^+, O^+, N^{2+}, and CO^+ are at the edge of, or outside, the stability region. Figure 2 shows the TOF mass spectrum of the C^{2+} ions released from the ion trap after 400 ms storage time with N_2 as the target gas in the reaction chamber. It can be seen that C^{2+} is well separated from N^{2+} in the TOF spectra, therefore the presence of N^{2+} in the trap did not affect the determination of C^{2+} ions for data analysis.

Each target gas—H_2, N_2, He, and CO—was admitted separately into the reaction chamber through a separate gas-handling system for each measurement. Each gas pressure was measured separately with a calibrated quadrupole mass analyzer. The absolute calibration of gas pressure has been discussed in a previous publication [17].

III. DATA ANALYSIS AND RESULTS

Figure 3(a) shows the C^{2+} ion signal intensity versus delay time plots for five different H_2 pressures. The scatter on
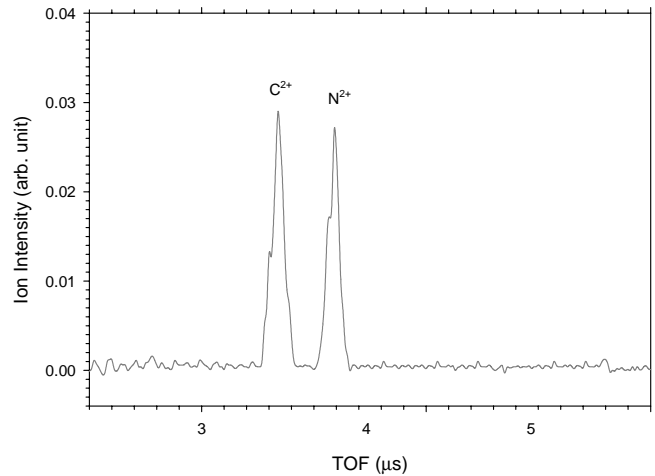

FIG. 2. TOF mass spectrum of the C^{2+} ions released from the trap after 0.4 s. N^{2+} is well separated from C^{2+}.
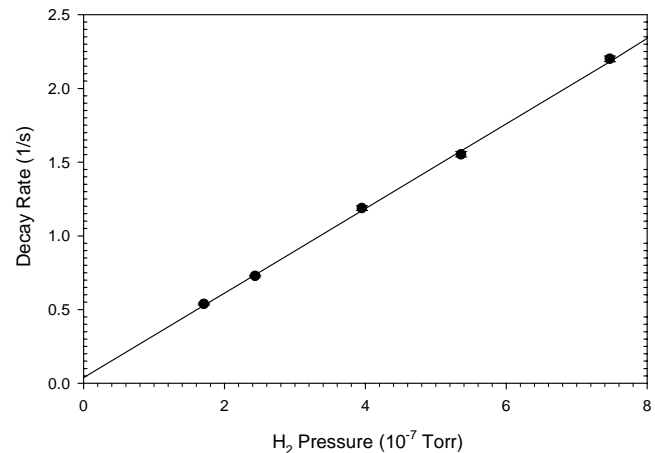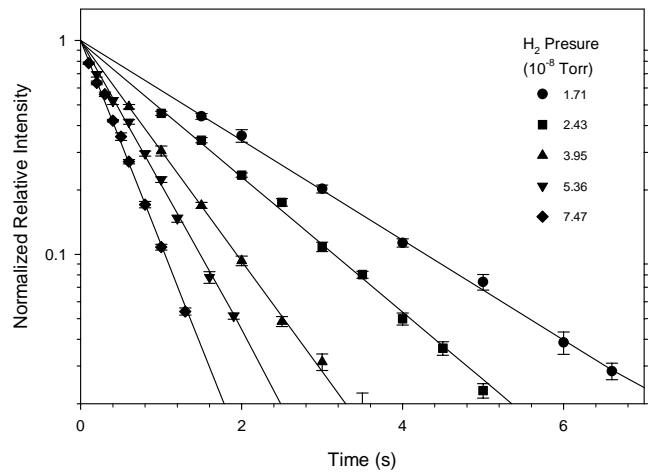

FIG. 3. (a) Decay curves of normalized relative intensity of C^{2+} ions vs storage time at different pressures of H_2 in a near-spherical pseudopotential well with $qD_z = 10.73$ eV and $qD_r = 12.68$ eV. The solid line is the least-squares fit to a single exponential function. (b) C^{2+} decay rate vs H_2 pressure.
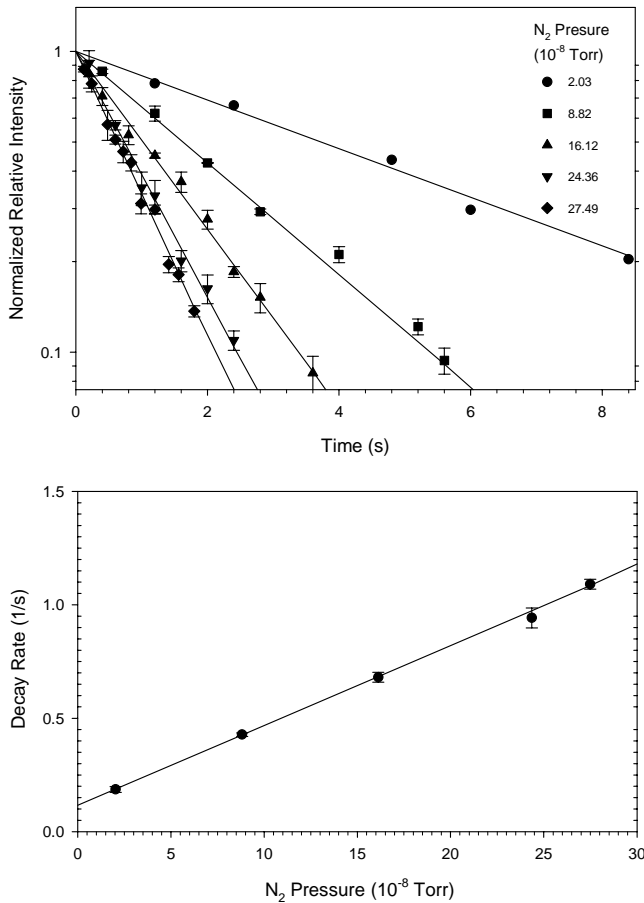
FIG. 4. (a) Decay curves of normalized relative intensity of C^{2+} ions vs storage time at five different pressures of N_2. The solid line is the least-squares fit to a single exponential function. (b) C^{2+} decay rate vs N_2 pressure.
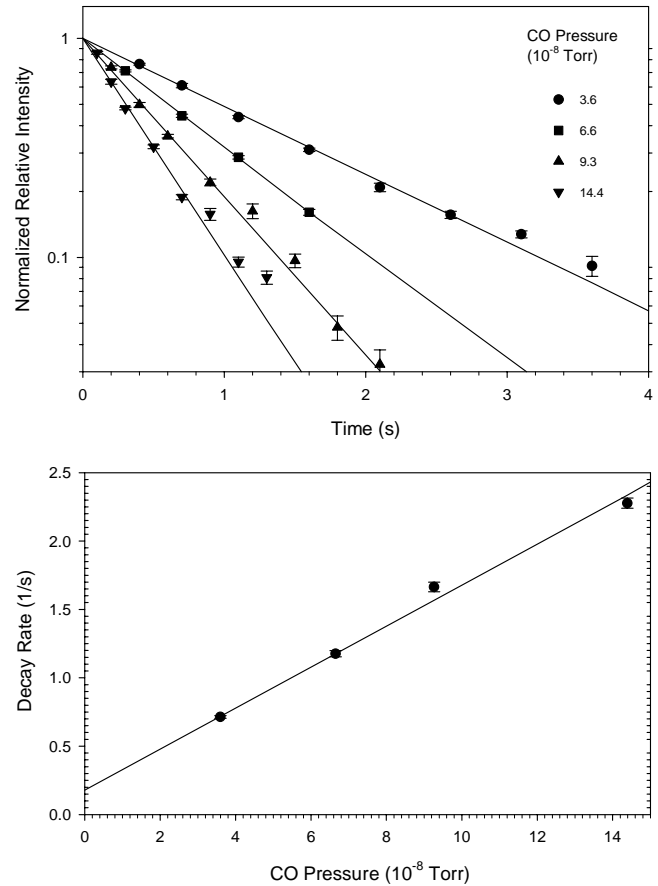

FIG. 5. (a) Decay curves of normalized relative intensity of C^{2+} ions vs storage time at five different pressures of CO. The solid line is the least-squares fit to a single exponential function. (b) C^{2+} decay rate vs CO pressure.

the data points is due to the fluctuation of the ion signals. The solid lines are the least-squares fit of the data by a single-exponential decay function. The slopes of the fitted lines in Fig. 3(a) give the charge-transfer rates at a given pressure of the target gas. The plot of the decay rates of the stored C^{2+} versus the pressure of H_2 gas is shown in Fig. 3(b). The slope in Fig. 3(b) is obtained by the weighted least-squares fit by a linear function, and the rate coefficient for C^{2+} interaction with H_2 is obtained from this slope. Similarly, the behavior of charge-transfer reactions rates for C^{2+} with N_2 and CO is shown in Figs. 4 and 5, respectively. The charge-transfer rate coefficients of C^{2+} are measured to be $8.77\,(\pm 0.71)\times 10^{-11}$ cm^3/s with H_2, $1.08\,(\pm 0.07)\times 10^{-10}$ cm^3/s with N_2, and $4.58\,(\pm 0.29)\times 10^{-10}$ cm^3/s with CO, respectively. The charge-transfer reaction rates for C^{2+} with H_2 and N_2 are similar, but both reactions are slower than that for C^{2+} on CO.

IV. DISCUSSION

Since the target gases H_2, N_2, and CO are at room temperature (300 K) and the temperature of the stored C^{2+} ions, corresponding to about $\frac{1}{10}$ of the pseudopotential well depth [21–23], is 1.7×10^4 K, an equivalent temperature T_{equiv} is

introduced to reflect the mean relative velocity of the C^{2+} and the H_2, N_2, and CO molecules. This equivalent temperature of the collisional system is given by

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

where the ion temperature is T_i, the target gas temperature is T_n, m_i and m_n are the masses of C^{2+} ion and target molecules, respectively, and μ is their reduced mass. The estimated equivalent temperatures are 2.63×10^3 K, 1.17×10^4 K, 1.17×10^4 K, and 4.38×10^3 K for H_2, N_2, CO, and He, respectively.

The C^{2+} is Be-like and has three low-lying metastable states, $2s2p\ ^3P_{0,1,2}$. Some fraction of the C^{2+} ions can be collisionally excited to the metastable state by the electron impact excitation [24,25]. The effect of the metastable ions in the charge-transfer measurements in the ion beam was observed by Unterreiter *et al.* [16] and Greenwood *et al.* [26]. About 10% of the ions were determined to be in the metastable state [16]. Recent calculation by Errea *et al.* [27] predicts that, at a collision energy of 600–10 000 eV, the cross section of electron capture by the C^{2+} ion from H_2 at a metastable state could be twice that at ground state. However, the amount of metastable state ions in the ion beam can be controlled by the electron impact energy. At an electron

impact energy of 70 eV, the amount of the ions at the metastable state was not observable [16].

The lifetime of the $2s2p\ ^3P_1^o$ metastable state was measured to be 8.3 ± 0.5 ms by Kwong *et al.* in this laboratory using electron impact excitation [25]. C^{2+} ions produced in the $2s2p\ ^3P_1^o$ state will likely decay to their ground state during the initial relaxation time of 400 ms built into the measurement timing protocol. However, the lifetime of the $2s2p\ ^3P_0^o$ and the $2s2p\ ^3P_2^o$ states has been predicted to be of 2×10^{10} s [28] and about 200 s [29], respectively. The C^{2+} ions produced at these metastable states are likely to stay in the trap and contribute to the measured rate coefficients. We checked the contribution by the metastable fraction in the current measurement by varying the electron impact excitation energy from below threshold energy ($V_{th} = 42.13$ eV) to more than three times the threshold energy, i.e., from $V = 41$ V to $V = 150$ V. We observed no difference in the measured rate coefficient within the experimental uncertainty. This suggests that the contribution by the metastable fraction is negligible.

Finally, the charge-transfer reaction rate of C^{2+} with He was measured. Since this measurement is extremely time-consuming due to the slow reaction rate, the rate coefficient was determined only to its upper limit. This determination is based on the C^{2+}—He reaction at two different pressures (i.e., 1.5×10^{-7} and 6.5×10^{-7} Torr). With the He pressure changes by a factor of 4, no observable difference was found in the storage time of the C^{2+} ion within experimental uncertainty. An upper limit on the charge-transfer rate coefficient is estimated to be less than 10^{-12} cm^3/s.

In summary, we have established the charge-transfer rate coefficients for C^{2+} with H_2, N_2, and CO to be 8.77 (± 0.71) $\times 10^{-11}$ cm^3/s (2.63×10^3 K), 1.08 (± 0.07) $\times 10^{-10}$ cm^3/s (1.17×10^4 K), and 4.58 (± 0.29) $\times 10^{-10}$ cm^3 s^{-1} (1.17×10^4 K). The rate coefficient for C^{2+} with He is estimated to be less than 10^{-12} cm^3/s (4.38×10^3 K). This confirms that the charge-transfer reaction of C^{2+} + He is a relatively slow process.

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