Electron-transfer collisions of low-energy multicharged nitrogen ions with H_2 and N_2

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A stored-ion collision technique has been used to study the charge-changing collisions of multicharged nitrogen ions with H_2 and N_2 target gases. The ions were produced inside a Penning trap with use of electron-impact ionization. The time constants for exponential ion loss by charge-changing collisions were measured at several target-gas densities, and from these time constants and densities the rate coefficients for electron capture were calculated. For the collision of N^{3+} with H_2, for which calculations by Gargaud and McCarroll were available, the cross section for electron capture was obtained from the rate coefficient using $\bar{v}$, where $\bar{v}$ is the rms ion velocity. The relationships of the other measurements to available theory and to ion-beam measurements at higher energies are discussed. Rate coefficients were obtained at mean ion energies near 1.1, 1.65, and 2.3 eV for N^{q+} charges q of 2, 3, and 4, respectively. The results of the rate coefficient determinations are $k(N^{2+},H_2)=[7.8(1.2)]\times 10^{-10}$ cm^3/s, $k(N^{3+},H_2)=[5.4(0.8)]\times 10^{-11}$ cm^3/s, $k(N^{4+},H_2)=[4.2(0.6)]\times 10^{-10}$ cm^3/s, $k(N^{2+},N_2)=[2.6(0.3)]\times 10^{-9}$ cm^3/s, $k(N^{3+},N_2)=[3.5(0.4)]\times 10^{-10}$ cm^3/s, and $k(N^{4+},N_2)=[2.0(0.3)]\times 10^{-10}$ cm^3/s.

I. INTRODUCTION

Cross sections or rate coefficients for electron transfer to various multicharged ions are important to the areas of research on x-ray lasers, fusion and laboratory plasmas, and ion sources, and in astrophysics. This wide-ranging need has led to substantial theoretical efforts[1] which at the lowest energies have principally been directed toward quantum-mechanical calculations of collisions of multicharged ions with atomic hydrogen. To a lesser extent, collisions of these ions with He and H_2 have also been studied, and a major effort devoted to more general approximations applicable to a wide range of collision partners at intermediate or higher energies.[2-4] At the lowest energies, the quantal theory involves the calculation of the orbitals and the potential-energy curves of incoming and outgoing states of the $(AH)^{q+}$ molecule, which is transiently formed by the collision of some general ion A^{q+} with H. Single electron transfer is then the only possible transfer process yielding bound electrons, although transfer with excitation can occur if the ion is not bare. The electron transfer is generally treated in terms of transitions induced by the nuclear motion between states of the same symmetry through radial coupling near avoided crossings of the adiabatic potential-energy curves.[5] Angular coupling is usually neglected at the lowest energies where it is small, and also because the avoided crossings most effective in electron transfer occur at relatively large internuclear separations.[1] An exact treatment requires the use of electron translation factors[1] to remove spurious couplings in the treatment of the collision, but for many states and at the lowest energies these are also often neglected in the calculations, or used only with approximations. When ions with several electrons are treated, some type of model potential[5] is used to deal with these additional electrons. Generally, in order to keep the calculations manageable, a truncated basis set is employed, chosen with the location and properties of the avoided crossings in mind. There seems to be an internuclear separation "reaction window"[6,7] in which, at a given velocity, avoided crossings are likely to lead to electron transfer, and outside of which the crossings are much less effective. When dealing with multielectron targets, the possibility of many-electron transfer makes the problem more complex.[8] It can be seen that the quantal theory applies largely to collisions with relative energies of a few electron volts or less, in which the approximations mentioned above are most valid, and the number of final states relatively limited.

Many experimental investigations of various collision partners have been completed, in part to provide desired information, and in part as a test of the theory. Most of these studies have been made using ion-beam technology,[9,10] necessarily performed at relative energies significantly higher than those to which the theory outlined above applies. Consequently there have been several attempts to extend the calculations to higher energies to compare theory with experiment.[11-13] At the higher energies, one or more of the approximations used in the theory may fail; it is desirable to provide experimental results in the lowest energy range, where the basic assumptions of the theory apply. An ion storage technique[14,15] permits the experimental study of multicharged ion-atom electron transfer collisions at very low energies, which are well within the domain of the quantal theory. Examples are the O^{2+}-H and O^{3+}-H collisions, which were recently studied using oxygen ions stored in a Penning trap, interacting with an atomic hydrogen beam.[16] The results of these measurements were in agreement with the calculations of Heil, Butler, Bienstock, and Dalgarno.[17,18] In this paper, we discuss the electron transfer collisions of nitrogen ions with H_2 and N_2 gases. These measurements are essential for planned studies with atomic hydrogen, but the N^{3+}-H_2 collision provides a direct and valuable test of a recent calculation

by Gargaud and McCarroll,[19] and the other measurements provide insight into the electron transfer collision process. In the following sections, after a brief description of the apparatus and technique, the individual collision studies are discussed in the light of theory and other measurements.

II. APPARATUS AND TECHNIQUE

The basic apparatus and techniques used in these measurements are relatively minor variants of those previously published,[15,20] and do not require exhaustive description. A Penning ion trap was installed in a bakeable ultrahigh-vacuum system pumped by a 60 l/s triode ion pump and a 250 l/s titanium sublimation pump. The base pressure of the system was 10^{-10} Torr following only a moderate bake. Target gases such as N_2 and H_2 were admitted to the system through a leak valve from a separately bakeable gas-handling system, which could be evacuated, after rough pumping, with a small ion pump. This system was used to mix gases, and to hold them at pressures near 1 Torr with minimal contamination.

The Penning ion trap was machined from molybdenum with surfaces which were hyperboloids of revolution having characteristic dimensions $z_0 = r_0/(2)^{1/2} = 0.6$ cm. The ring electrode was divided into two segments to permit efficient radial excitation of the stored ions. During measurements, the ions were confined by an axial magnetic field near 0.7 T, and by a negative dc potential $V = 11.5$ V applied to the trap ring electrode. Nitrogen ions were produced by electron-impact ionization of N_2 gas, typically held at a pressure of 2.5×10^{-8} Torr when H_2 collisions were studied. The ionizing electron current was typically 2 μA at a potential of 150 V.

To detect the ions, the axial oscillation frequency $\omega_z = (qV/mz_0^2)^{1/2}$ of each ion charge-to-mass ratio q/m was swept through resonance with an excited tuned circuit having a Q of 80, connected between the end caps of the trap. The ions absorbed rf energy, producing an amplitude modulation on the rf envelope proportional to ion number. The rf was amplified, detected, and the signals converted to pulses by a V/f converter, and then stored and integrated with a multichannel analyzer as a function of channel number correlated to the confining potential ramp (see Fig. 1). Undesired ions were removed from the trap with excitation at either the shifted cyclotron frequency $\omega_+ \simeq \omega_c - \omega_-$, where $\omega_c = qB/m$, or the magnetron frequency ω_-, driven by a computer-controlled synthesizer through rf transformers connected across the split ring of the trap.

Two types of measurement suffice to determine the rate coefficient for electron transfer from a target gas to a particular ion charge state. One is a measurement of the target-gas pressure, from which the density is obtained by a standard conversion. The gas pressure was determined with a nude ion gauge, calibrated at pressures greater than 10^{-5} Torr against a capacitance manometer. Readings were corrected for known gas sensitivities. Measurements of certain known rate coefficients, made by ion storage or other techniques, have been repeated, and used as a check on the density
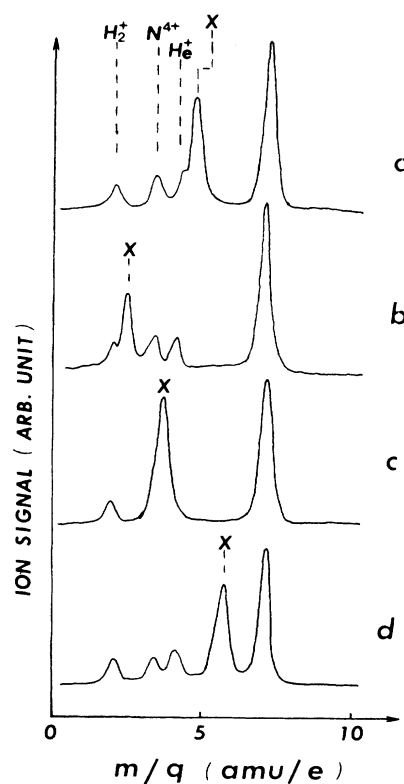

FIG. 1. Ion signals observed using the axial detection system, plotted vs m/q. (a) Signals showing the relative location of peak X, which was unidentified by cyclotron resonance. (b)-(d) Signals showing the relative location of peak X as the magnetic field was increased. Peak X is identified as a parametric excitation of the shifted cyclotron frequency ω_+ of stored N^+.

calibration. A determination of the time constant for ion loss due to charge-changing collisions with the target gas is the second basic measurement required. The ions were produced by an electron pulse, stored for a measured interval at constant trapping potential, and then detected. Repetition of these measurements at different storage times provided a plot of remaining stored-ion number versus storage time. When no metastable ions are found to be present, as in these measurements, the ion number decay is found to be exponential; the time constant τ for this decay is related to the rate coefficient for electron transfer k by $k = (n\tau)^{-1}$. When possible, τ is measured at several target densities n. A plot of τ^{-1} versus n extrapolates to the reciprocal time constant for electron transfer with any residual gas, and the slope is a measure of k for the target gas.

In general it was found to be difficult to optimize the ion signal of all nitrogen ion charge-to-mass ratios simultaneously. The signals depend on the electron current, the pressure, and the number and q/m of other ions confined simultaneously in the trap. With the chosen operating conditions, N_2^+ ions were not stably confined, but other ions such as N^+, which were formed in significant numbers due to large production cross sections and low loss rates in the target gas, were greatly re-

duced in number or eliminated by either ω_+ or ω_- resonant excitation. At low N_2 pressures, signals from residual gases such as He^+ or H_2^+ were relatively prominent, and these ions were removed. Generally N^{2+} ions were not totally removed since these ions were important to the production of the higher charge states N^{3+} and N^{4+}, demonstrating that N^{2+} ionization was a significant source of these higher charges.

Normally each charge state signal was identified not only by its position in the detected spectrum of peaks, in which signal position is proportional to the mass-to-charge ratio m/q, but also by direct cyclotron excitation which reduced the detected signal when at resonance. Figure 1 shows a sequence of ion signals detected by the usual axial excitation, without cyclotron excitation removal of any q/m states. In spectrum (a) the peaks due to N^{2+}, He^+, and H_2^+ can be readily identified. The peak near $m/q = 3$ is composed of H_3^+, produced by the reaction $H_2^+ + H_2 \rightarrow H_3^+ + H$, and possibly a small amount of N^{4+}. The large peak labeled X is in the proper location to be N^{3+}; however, it was not possible to remove this peak using the cyclotron frequency appropriate to N^{3+}. Instead, the peak was reduced at the cyclotron frequency of N^+. In the remaining sequence of signals (b)–(d) the magnitude of the confining magnetic field B has been incrementally increased, starting at a lower value. The signal X is seen to move relative to the other signals, which must have positions independent of B, since the axial oscillation $\omega_{zj} = (q_j V/m_j z_0^2)^{1/2}$. The peak X is in fact the shifted cyclotron frequency ω_+ of N^+, excited parametrically by a time-varying component of the electric field applied at ω_z between the two trap end caps with resonance at $\omega_+ = 2\omega_z$. The B dependence is shown in Fig. 2.

The mean energies of the individually measured stored ions are usually assumed to be approximately one-tenth of the energy of the potential well in which they are confined, based on experience with singly charged ions. Since the rate coefficient measurements were carried out
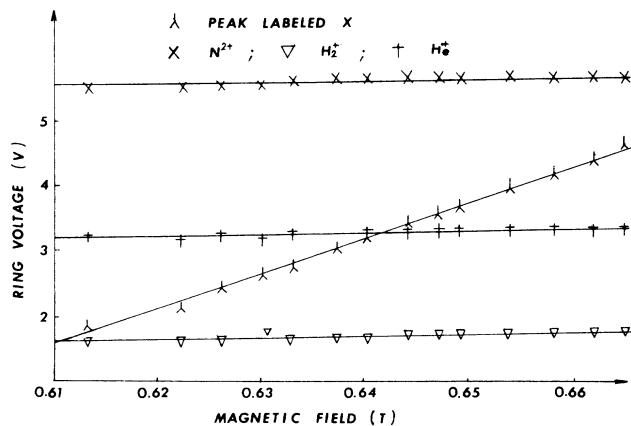

FIG. 2. Plot of the potential for appearance on the detection sweep of peak X vs the confining magnetic field. The apparent linear dependence on B is an approximation to the actual functional form. The independence on B of other signals is also shown.

separately for each charge state, there is no reason to expect a common energy for all nitrogen charge states due, for example, to ion-ion collisions. Attempts to approximately verify the mean energy of the relatively large number of N^+ ions were made by comparing two signals. The ion "number" signal was first obtained, as described above. The tuned circuit exciting rf was then switched off, and the gain of the amplifiers increased, to obtain a signal due to the incoherent motion of the ions in the trap, which induce current fluctuations in the tuned circuit. This signal is proportional to both ion number and mean ion energy. Using measured parameters, a calculation of the mean energy of stored N^+ ions gave a result near one-tenth of the energy of the storage well, as expected, with an uncertainty of about $\pm 50\%$. For N^{2+}, this would correspond to a mean center-of-mass energy near 0.1 eV/amu, an estimate expected to be comparably accurate. With similar relative uncertainties for the mean ion energy of the N^{3+} and N^{4+} charge states, a value of one-tenth of the storage well energy for each charge state has been chosen. Attempts to observe random current fluctuations for the higher charge states were unsuccessful, which, however, may indicate a lower mean energy than this estimate.

III. RESULTS AND DISCUSSION

First the rate coefficients for the nitrogen-ion charge states in nitrogen target gas were measured, and then similar determinations of the rate coefficients of the same ion charge states in known mixtures of N_2 and H_2 gases were made. Since more theoretical information is available about electron transfer with target H_2, these measurements will be described first, followed by the measurements in N_2. In all cases, the H_2 measurements were performed with the same pressure of N_2 gas in the vacuum system.

A. N^{2+}-H_2

The measurements reported here are the first results for this electron transfer interaction at electron volt energies. Total cross sections for electron transfer have been obtained recently at ion-beam energies between 600 eV and 200 keV by Wilkie et al.[21] and an energy-charge spectrum measured at 8 keV by McCullough et al.[22] Earlier cross-section measurements at comparable energies by Flaks and Ogurtsov[23] and by Phaneuf et al.[24] yielded higher values, due apparently to contamination of their N^{2+}-ion beams by metastable excited ions.[21] The cross sections for incident $N^{2+}(2s2p^2)^4P$ metastable ions are approximately two times larger than those for $N^{2+}(2s^22p)^2P^\circ$ ground-state ions with the same energies.[21] Figure 3 shows the single-exponential decay of our N^{2+} sample at several target pressures of N_2, and hence at different time scales relative to the fixed mean life of the metastable level. Similar results were obtained in H_2.

There are no calculations for the N^{2+}-H_2 collision, although the N^{2+}-H charge transfer collision has been treated by Heil et al.[25] below 8 eV and subsequently up
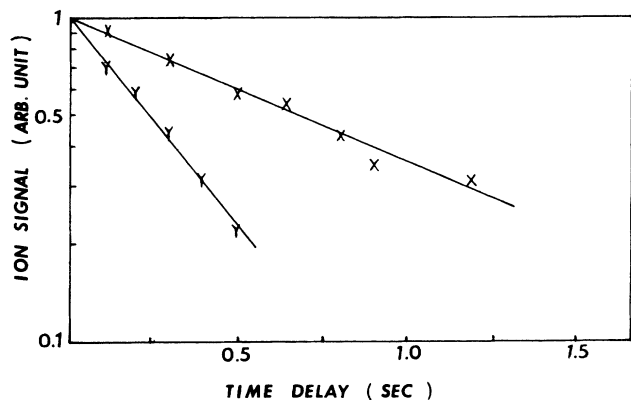
FIG. 3. Plots of N^{2+} ion number vs storage time at 10^{-8} Torr (x) and 3×10^{-8} Torr (y) pressures of target N_2 gas, showing single-exponential decrease due to reactions.

to 5.4 keV/amu. A single product channel, $N^+(2s 2p^3)^3 D^\circ + H^+$ was included, which involved electron excitation as well as transfer, but was still expected to be dominant.[26] Near 0.11 eV/amu the cross section was calculated to be 16.6×10^{-16} cm^2. Based on the 4.58-eV exothermicity for this final state, which was observed by McCullough et al.,[22] the crossing radius R_{xH} is 6 a.u., a number very close to the result obtained by assuming that $\sigma = \pi R_{xH}^2$. Due to the 1.8-eV difference in ionization potentials between H_2 and H, the exothermicity for formation of $N^+(2s 2p^3)^3 D^\circ + H_2^+(^2\Sigma_g^+ (v=0))$ should be 2.75 eV, corresponding to a crossing radius near 10 a.u. This, plus transfer to higher vibrational levels, was observed among other channels by McCullough et al.[22] at 8 keV. The vibrational levels of H_2^+ are separated by about 0.27 eV,[27] leading to avoided crossings at larger radii, which will have a lower probability of excitation. Consequently, we assume that near 0.1 eV/amu energy, only the $v=0$ level of the H_2^+ $^2\Sigma_g^+$ state will be populated, i.e., that a single channel (in addition to the incoming channel) is probably adequate to describe the collision. This is consistent with the N^{3+}-H_2 collision data discussed below.

Metastable levels of the stored N^{2+} ions would lead to ion loss curves characterized by two time constants, associated with the two different rate coefficients, had such metastables been present. Analysis of the observed single-exponential decays measured at five different pressures, as shown in Fig. 4, yielded a rate coefficient for the N^{2+}-H_2 collision $k(N^{2+},H_2) = 7.8(1.2) \times 10^{-10}$ cm^3/s at a mean ion energy near 1.1 eV. This corresponds to a mean electron transfer cross section $\sigma \simeq 20 \times 10^{-16}$ cm^2, which is approximately 20% higher than the calculated cross section at the same energy for the N^{2+}-H collision. At the higher energies studied by Wilkie et al.,[21] the calculated cross sections for N^{2+}-H electron transfer were about 20% higher than experiment in the energy range where the single reaction channel approximation was valid. For avoided crossings at radii between 6 and 10-11 a.u., the cross sections and reaction rates for collisions of H and H_2 can be expected to be comparable, if they are near the theoretical max-
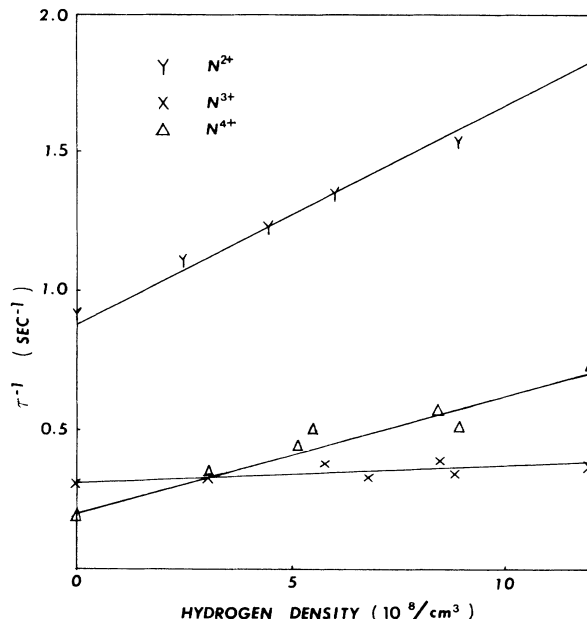

FIG. 4. Plots of reciprocal measured nitrogen-ion storage time constants vs the density of H_2 gas. The time constants at zero H_2 density were measured in the constant background of N_2 gas, from which the nitrogen ions were produced. Charge changing with residual gas was negligible. The rate coefficients for charge changing were determined from least-squares fits to these data.

imum, as pointed out by Gargaud and McCarroll[19] for the C^{4+}-H and C^{4+}-H_2 collisions. This seems to be the case also for N^{2+}-H and N^{2+}-H_2.

B. N^{3+}-H_2

It has been possible to study the N^{3+}-H electron transfer collision theoretically since the ^1S core of N^{3+} is ideally suited to model potential methods. Gargaud and McCarroll point out that the N^{3+}-H collision, in which capture into the $N^{2+}(1s^2 2s^2 3s)^2S$ state occurs via a crossing at 8 a.u., can be expected to proceed at a rate near the theoretical maximum, where the accuracy of the theory can be expected to be high. The N^{3+}-H_2 localized crossing near 13.2 a.u. provides a much more stringent test of the theory, since at higher velocities the crossing appears diabatic and the cross section becomes quite small. At the lowest energies, which are of interest here, the calculated cross sections for N^{3+}-H and N^{3+}-H_2 appear nearly identical, as noted above for the case for N^{2+}. Linear extrapolation of the N^{3+}-H cross-section plot in Fig. 11 of Ref. 19 to 0.1 eV/amu yields a cross section near 1×10^{-16} cm^2, compared to 9×10^{-17} cm^2 at 0.1 eV/amu obtained directly from Fig. 12 of Ref. 19 for the N^{3+}-H_2 interaction.

Due to the small value of the electron transfer rate coefficient, the N^{3+}-H_2 measurements were carried out at seven H_2 pressures above the constant base pressure of 2.5×10^{-8} Torr of N_2. The transfer rate coefficient for N^{3+} in N_2 is nearly six times larger than that for this ion in H_2. Figure 4 shows a plot of the reciprocals of

the measured ion storage time constants versus the density of H_2 gas. A least-squares fit to the data yields a slope $k = [5.4(0.8)] \times 10^{-11}$ cm^3/s. Using the assumed maximum stored-ion energy of 1.65 eV to obtain the mean ion velocity, the mean cross section is $\sigma = [9.3(1.4)] \times 10^{-17}$ cm^2. This result is plotted in Fig. 5 with the theoretical curve;[19] it differs from the preliminary result[28] largely in the improved data analysis, which has enabled a reduction in the uncertainty. The agreement with theory implies that the approximations used by Gargaud and McCarroll, namely, neglect of electron translation factors and angular coupling, and the use of model potentials for N^{3+} and H_2, are appropriate for this collision system at very low energies. Since this system has a single theoretical reaction channel, the agreement with experiment implies also that the higher vibrational levels of H_2^+ are not excited. The large disagreements of the theory with experiment at higher energies[21,29] are clearly due to the opening up of channels not treated in the theory.

C. N^{4+}-H_2

N^{4+} ions change charge slowly in N_2 gas, at about half of the rate compared to N^{4+} in H_2. For this ion, with a $(1s^2 2s)^2 S_{1/2}$ ground state, there are no theoretical calculations for collisions with either H or H_2, presumably because of the difficulty in modeling the electrons bound to the ion. The mean ion energy was estimated to be 2.2 eV or less in these measurements. A plot of τ^{-1} versus H_2 density is shown in Fig. 4. Data at much higher collision energies show a larger cross section for capture, which is velocity independent.[30] The rate coefficient is $k(N^{4+}, H_2) = [4.2(0.6)] \times 10^{-10}$ cm^3/s. The rates for charge transfer in H_2 are summarized in Table I.
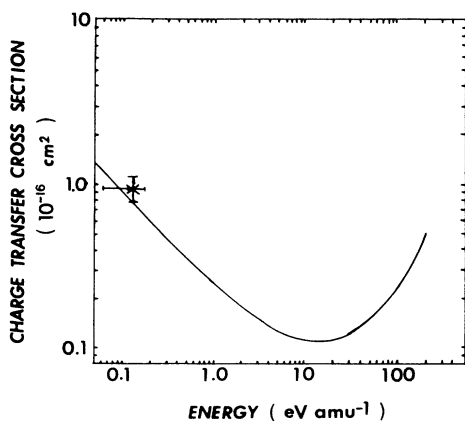

FIG. 5. Plot of theoretical cross sections for electron transfer from H_2 to N^{3+} (solid line) from Fig. 12 of Ref. 19, and the experimental cross-section point determined from the measured rate coefficient divided by the rms ion velocity. The horizontal scale is the center-of-mass energy divided by the reduced mass.

TABLE I. Rate coefficients k (units of cm^3/s) for electron transfer from H_2 and N_2 to N^{q+} ions measured near 1.15, 1.65, and 2.3 eV mean energies of charge states $q = 2$, 3, and 4, respectively.

	H_2	N_2
N^{2+}	$[7.8(1.2)] \times 10^{-10}$	$[2.6(0.3)] \times 10^{-9}$
N^{3+}	$[5.4(0.8)] \times 10^{-11}$	$[3.5(0.4)] \times 10^{-10}$
N^{4+}	$[4.2(0.6)] \times 10^{-10}$	$[2.0(0.3)] \times 10^{-10}$

D. N^{q+}-N_2 collisions

There are no quantum-mechanical theories for electron volt energy electron transfer collisions of multicharged nitrogen ions with N_2, so these measurements provide the only available information on these interactions. These rates are important in verifying and establishing the background rate for the H_2 collision data, since N_2 gas must be present in the measurements to supply a source of ions. The reciprocal lifetime in the N_2 gas appears in Fig. 4 as the data point in the absence of H_2; it serves a significant role in the least-squares fits to the H_2 collision data. The rate coefficients for the N_2 collisions, listed in Table I, decrease in magnitude as the ion charge state increases, in direct contradiction to the predictions of classical Langevin theory, in which the rate coefficient or cross section scales as q; $k_L = 2\pi q e (\alpha / \mu)^{1/2}$. Using $\alpha(N_2) = 1.7 \times 10^{-24}$ cm^3,[31] $k_L(N^{2+}, N_2) = 2 \times 10^{-9}$ cm^3/s, which is smaller than the measurement. The rates for the higher charge states are smaller than the Langevin rates by factors of 10 and 20, respectively. The measured rate coefficient $k(N^{2+}, N_2)$ was obtained in a previous ion storage measurement, with lower precision; the collision is discussed in detail in Ref. 14.

The ionization energy of N_2 is nearly the same as that of H_2, so the avoided crossings of the potential-energy curves may not differ significantly from the N^{q+}-H_2 crossing positions. Indeed, the trend of rate coefficient magnitude from N^{2+} to N^{3+} is the same for H_2 and N_2 targets, but the N^{4+} rate in N_2 remains low, in contrast to the rate in H_2. The magnitudes of k are small in both gases, however.

IV. CONCLUSION

The rate coefficients for electron volt energy electron transfer collisions of N^{q+} ions $(2 \leq q \leq 4)$ with H_2 and N_2 targets have been measured using a stored-ion collision technique. The N^{3+}-H_2 rate data, when converted to a cross section, are in good agreement with the theoretical calculations of Gargaud and McCarroll. Other rates are discussed in terms of the theory of collisions with atomic hydrogen, and with ion-beam experiments at higher energies. Studies of low-energy multicharged ion charge-changing collisions using ion traps continue to be an effective means for obtaining new information and testing theoretical calculations.

ACKNOWLEDGMENTS

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