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Charge-Transfer Cross Sections for H^+, Li^+, and Na^+ on N_2 †

Grant J. Lockwood
Sandia Laboratories, Albuquerque, New Mexico 87115
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Measurements of the total cross section for charge transfer σ_{10} of H^+, Li^+, and Na^+ on N_2 are reported here in the energy range of 25–100 keV. The method used was direct detection of fast neutral particles that were formed in single charge-transfer collisions. Data for H^+ on N_2 are in excellent agreement with previously published results with σ_{10} increasing with decreasing energy, reaching a value of 1.55×10^{-15} cm^2/molecule at 5 keV. From data for Li^+ and Na^+ on N_2, σ_{10} is observed to increase with increasing energy. The cross section for Na^+ has a maximum of 1.1×10^{-16} cm^2/molecule at about 50 keV, while the cross section for Li^+ is still increasing at 100 keV, although an irregularity is observed at approximately 30 keV.

I. INTRODUCTION

Measurements have been reported of the total cross sections for charge transfer of Li^+ and Na^+ on N_2 by Ogurtsov *et al.*[1] in the energy range ~1–30 keV by detecting the fast neutral particles formed in single charge-transfer collisions. This method has been used in the present study which extends the energy range to 100 keV. In addition, the charge-transfer cross section for H^+ on N_2 by this method is included. This result is compared with data reported by other investigators[2–4] using other techniques.

The method's main shortcoming is that neutrals scattered through an angle larger than 1° are not detected. This can cause the value of the cross section to be low at low energy, but in the energy range covered here this does not appear to be the case. Jones *et al.*[5] have measured the contribution to the total cross section for particles scattered between 0° and 1°. For charge transfer of noble-gas ions in noble gases, at 25 keV all cases showed that 96% or more of the total cross section was contained between 0° and 1°, with the exception of Ne^+ on Ar where only 85% was measured between 0° and 1°.

Section II describes the apparatus and procedure used to obtain the data. Data are presented and discussed in Sec. III, and comparisons are made with data of other investigators.[3,6]

II. APPARATUS AND EXPERIMENTAL PROCEDURE

The Sandia Laboratories 100-kV ion accelerator was used in this study. The H^+ ions were obtained from an rf ion source of the Oak Ridge type, whereas the Li^+ and Na^+ ions were from zeolite sources.[7,8] The choice of ion source was governed by the need to prevent formation of metastable ions in the incident beam. The cross sections for charge transfer to ground-state and metastable ions would be different, and a beam of unknown mixture would produce a cross section to which theoretical or experimental comparisons would be meaningless. Since H^+ has no excited states, the rf source was used, but it was not used for the Li^+ or Na^+ as described in previous studies.[9] Although the exact mechanism by which the zeolite sources produce ions is not understood, it is believed that most of the ions must be in the ground state. This is based on the fact that these sources are thermionic ion emitters and thus cannot produce metastables. Excitation to metastable levels, which lie at 59.0 eV for Li^+ and 32.8 eV for Na^+, by impact with secondary electrons created in the source is insignificant. The accelerator delivers a beam of magnetically analyzed ions having small energy spread to the experimental chamber. The energy is measured to 2%.

The experimental apparatus is shown in Fig. 1. The incident ion beam, collimated to ±0.10° by apertures A and B, enters the collision chamber through B. Immediately behind B inside the collision chamber is a movable monitor, M. After traversing 0.91 cm of target gas, the beam exits from the collision chamber through hole C. Holes C and D are used to collimate the scattered beam and allow all particles which have been scattered into a cone of half-angle 1.0° to reach the detector, while ensuring that no scattered particles can hit the electrostatic analyzer behind hole D. Following the electrostatic analyzer is a secondary electron detector which is used to measure both the incident ion beam and the neutral beam produced by charge transfer.

To make certain that holes C and D were centered on the beam axis as determined by holes A and B, that part of the system containing holes C and D, the electrostatic analyzer, and the detector, were pivoted about point P; current to the detector was recorded as a function of angle on both sides of the beam axis. From a plot of current versus angle, the 0° direction was determined and the movable portion of the system set and locked at 0°. During this process, the incident beam marked the aperture containing hole C when the angle was large enough to cut off the incident beam. This marking was visually checked to determine that the system was in vertical alignment. These checks were performed in addition to the mechanical alignment which located the centers of all elements to within 0.025 mm of the axis determined by A and B.

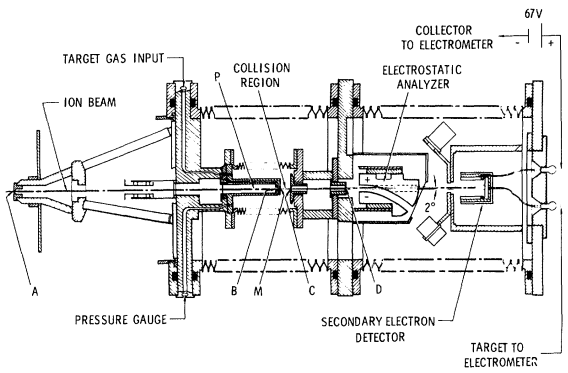

FIG. 1. Experimental apparatus.

The cross sections reported here were obtained from the equation

$$\sigma_{10} = N_0/N_1 ml\,, \tag{1}$$

where N_0 is the number of neutrals in the scattered beam, N_1 is the number of incident ions, m is the number of target molecules per unit volume, and l is the thickness of the target in the direction of the incident ion beam.

For these measurements, N_0 and N_1 were not determined directly, but a current associated with them was measured. The detector used was of the secondary electron type. The target was held at ground potential while the collector was maintained at +67 V with respect to the target. This voltage was sufficient to ensure saturation of collector current at all ion energies. Currents to both the target and the collector were recorded. The collector current I_C is given by

$$I_C = N\gamma\,, \tag{2}$$

where N is the number of particles hitting the target and γ is the number of secondary electrons ejected from the target per incident particle. In the energy range covered by this study, γ is, to a good approximation, independent of particle charge for a fixed particle energy.[10,11] However, γ could still depend on surface conditions and might vary with gas pressure or beam current. To determine if either of these conditions might affect the data, preliminary experiments were conducted. Data were taken with a target heated sufficiently to keep an N_2 layer from forming, and γ was measured for a wide range of ion beam currents by defocusing the beam. Results of both tests led to the conclusion that surface conditions would not affect the data. Furthermore, such a change in γ caused by surface conditions would

result in a nonlinear relationship between I_0 and pressure, which is precluded by results shown in Fig. 2. Thus, Eq. (1) can be rewritten in terms of known constants and quantities actually measured in the laboratory as

$$\sigma_{10} = (I_0/I_1)\ T\ 1.035 \times 10^{-19}/Pl, \qquad (3)$$

where I_1 is the collector current when the incident ion beam was allowed to strike the target, I_0 is the collector current when all charged particles in the scattered beam were swept away by the electrostatic analyzer and only the neutral components reached the detector, T is temperature in °K, and P is pressure in Torr. Because a single detector was used to measure both currents an absolute determination of γ was not necessary, since it cancels when the ratio of the currents is taken within the above approximation.

In obtaining measurements of the variables in Eq. (3), certain experimental conditions must be considered. First, the incident beam may have a neutral component because of charge transfer in the accelerator, after the magnet. Second, when the target gas is turned off, there is residual gas in the collision chamber with which the incident ion beam can effect charge transfer. Finally, the actual thickness of the target gas may be greater than the physical dimension between holes B and C, caused by the effusion of target gas through these holes. The data were obtained in such a way that the first two effects were automatically minimized. In the procedure used, the ratio I_0/I_1 was obtained for at least five different pressures in the range from 2×10^{-4} to 12×10^{-4} Torr at each energy. A plot of I_0/I_1 versus pressure was made from the data and the cross sections determined from the slope of the line obtained by a least-squares fit to the data. In Fig. 2, which is a typical plot of this type, it can be observed that the line through the data points does not go through zero when extrapolated to zero pressure. This positive value of the ratio I_0/I_1 can come from the first two experimental conditions mentioned above or from an offset in the pressure gauge. Since it is the slope of the line that is used in determining σ_{10} and not the individual points, this effect makes no contribution to the calculations of σ_{10}. This procedure also relaxes the requirements on pressure measurements. It is only necessary that the pressure measuring system be linear over a small range. A small offset in the absolute pressure calibration will not affect the calculated value of σ_{10}. However, an improper scale factor would result in error. As seen from Fig. 2, the plot of the data is indeed linear in the pressure range used. This is expected[5] since only single collisions should occur in this pressure range. It therefore follows that either the pressure measuring system is linear or that both it and the data are nonlinear in a compensating manner. The latter seems unlikely, however. The pressure measurements were obtained with a Baratron gauge. The manufacturer's calibration was used and is stated to be better than 0.1% absolute, as used.

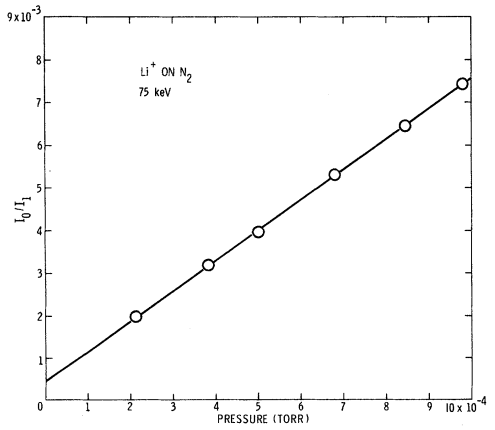

FIG. 2. Ratio of current associated with the neutral particles to current associated with the incident ion beam versus target gas pressure for single collisions of Li^+ on N_2 at 75 keV. The slope of the line which is obtained by a least-squares fit to the data is used in determining the σ_{10} cross section.

The actual thickness of the target gas in the direction of the incident ion beam can be affected by two conditions associated with the effusion of gas through holes B and C. First is the problem of the gas immediately outside the holes. From effusion relations, it is found that the number density of molecules integrated over the path length will constitute approximately a 1% correction to the target length as measured by the distance between holes B and C. This correction is neglected in the present measurements. The second problem is concerned with the increase in over-all system pressure while the target gas is on, because of the long path length of beam outside the collision chamber. In the present case, the system pressure was maintained at 10^{-3} times the chamber pressure and thus presented no problem.

III. DATA

Data from the present study are presented in Figs. 3–5, together with data from other investigators.[1–4,6] A measure by which the present data may be compared is the adiabatic criterion by Massey.[12] For charge transfer between unlike particles, the criterion predicts that the cross section should be small at low velocity, rise to a maximum, and fall as the velocity is increased above that for the maximum. The main

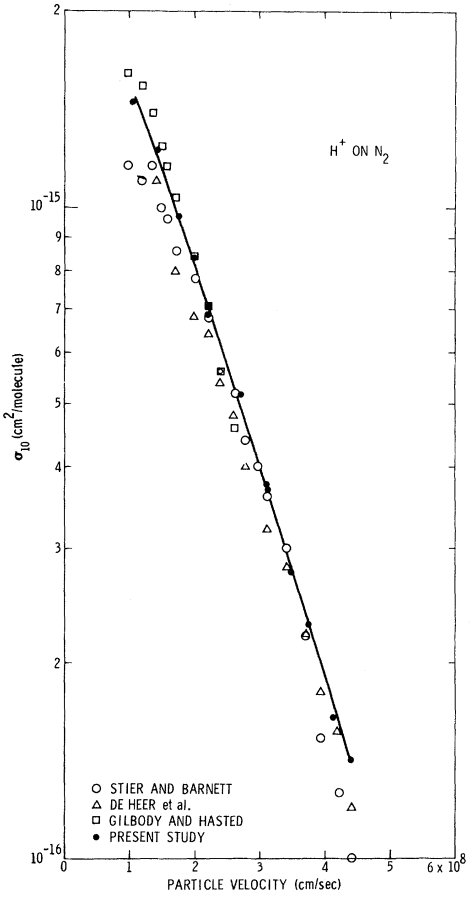

FIG. 3. Total cross section for charge transfer σ_{10} of H^+ on N_2 versus velocity of the incident H^+.

problem is using the adiabatic criterion for prediction of the velocity for the maximum in the cross section is the choice of ΔE, the internal energy defect. It is evident from analysis of experimental data[13] that the energy defect may not be simply that at infinite nuclear separation, but, in the present study, might be dominated by polarization forces. Further, the role of excited states in this process may be important in some cases. Despite the inability to obtain quantitative comparisons with the criterion, qualitative results may be examined.

For the case of H^+ on N_2, present data show the cross section decreasing with increasing velocity. This is as expected, if the maximum in the cross section is at a velocity below the minimum reached by the present study. As can be noted in Fig. 3, agreement between present results and those of the other three investigators[2-4] is excellent. If a mean curve is drawn for all the data, most experimental points fall within a band of ±10%. For the combination H^+ and N_2, the data were extended down to 5 keV to obtain at least a lower limit for

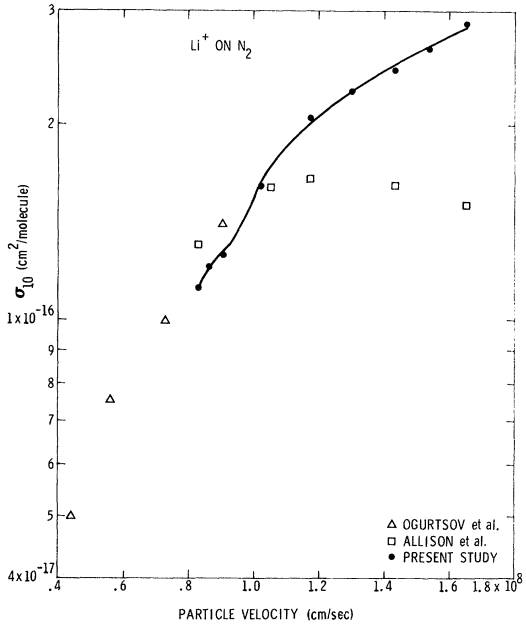

FIG. 4. The total cross section for charge transfer σ_{10} of Li^+ on N_2 versus velocity of the incident Li^+.

the cross section, depending on large-angle scattering. Data at the lower energies do not show any tendency to fall and, in fact, are above those of two investigators.[2,4] This is taken as evidence in the case of H^+ on N_2 that large-angle scatter is not important down to 5 keV.

Figure 4, in which present data for Li^+ on N_2 are compared with those of two other investigators[1,6] does not show the excellent agreement that H^+ on N_2 did. In the very limited velocity range (8×10^7

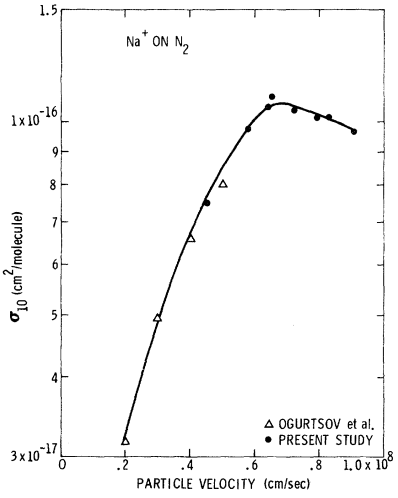

FIG. 5. The total cross section for charge transfer σ_{10} of Na^+ on N_2 versus velocity of the incident Na^+.

to 11×10^7 cm/sec), where all three have data, the agreement is very good, but at higher velocity the present data are well above those of Allison,[6] reaching a factor of 2 at 1.65×10^8 cm/sec. The low value obtained by Allison, using charge-equilibrium methods, can be attributed to two sources. First, small-angle scattering of neutrals from the beam is very important because of the converter cell geometry used. Particles scattered through an angle $>0.13°$ at the entrance and $>0.23°$ at the midpoint will not be detected. Measured angular distributions of light atom scattering indicate that approximately 10% of the particles are found in the region between $0.16°$ and $1.0°$, at an incident velocity of 1.55×10^8 cm/sec. The rise in a constituent with increased pressure does not ensure that many particles are not being lost through small-angle scattering, but simply that the production rate at higher pressure is greater than the loss. Second, the equations used to represent the condition of the equilibrated beam contain single-value cross sections and neglect charge-transfer cross sections from excited states, which can be important at the higher pressures where the time between collisions is short. Furthermore, if the present measurements were affected by small-angle scattering (which is not believed to be the case) the cross sections would be too small and, since they lie above those of Allison,[6] it is believed the Allison-deduced cross sections are low by a factor of 2 or more at the highest energy used in this study.

Present data for Na^+ are shown in Fig. 5, where Ogurtsov's[1] data are also shown. Although the overlap in velocity is small between the two studies, a comparison can be made. It is noted that the sets of data do match well where they meet, and a single smooth curve can be drawn through all the data. Data show a maximum in the σ_{10} cross section at a velocity of 6.5×10^7 cm/sec. With reference to the adiabatic criterion, if the ΔE at infinite separation were usable, the maximum in the Na^+ on the N_2 and Li^+ on N_2 cross sections would be expected to be close to the same velocity, since the respective ΔE's of 10.46 eV for Na^+ on N_2 and 10.21 eV for Li^+ on N_2 differ only by 0.25 eV. As can be observed in Fig. 4, the σ_{10} cross section for Li^+ on N_2 is still rising at 16.5×10^7 cm/sec.

IV. CONCLUSIONS

Although discussion of the results given above is applicable to the general shape of the σ_{10} curves, it should be noted that there is evidence from the present experiment of an irregularity in the cross section for Li^+ on N_2 at about 8.5×10^7 cm/sec. The curve in Fig. 4 could be fitted by a smooth structureless curve within the stated error. However, many data points were taken in the velocity region around 8.5×10^{-7} cm/sec and the data were found to scatter widely, while data taken on either side of this region were well behaved. The points

TABLE I. Sources and estimated uncertainties.

Source	Uncertainty (%)	Remarks
Target thickness, l	1	See discussion, Sec. II.
Neutral current reading, I_0	3	Electrometer limitation.
Ion current reading, I_1	3	Electrometer limitation.
γ cancellation in ratio I_0/I_1	5	Based on laboratory studies, including effects of surface temperature, target material, and current density striking target.
Pressure, P	2	Maximum estimated error due to scale factor. This is conservative, based on manufacturer's specifications.
Small-angle scattering	5	Based on results given in Ref. 5 and laboratory studies with H^+.

plotted are an average of the many points taken. This could indicate that above 8.5×10^7 cm/sec a different ΔE is applicable, while below, a ΔE close to the ground-state energy difference at infinite separation, ΔE_g, applies. There are many possibilities in the complicated N_2 system which could account for a ΔE larger than ΔE_g. From electron impact studies,[14] appearance potentials for N_2^+ have been measured at 15.6, 16.9, 18.8, and 23.6 eV. Furthermore, the state of the target after the collision is unknown; thus dissociation of the N_2 can not be ruled out. If indeed, dissociation and ionization are occuring at higher velocities, appearance potentials of 24.3, 26.2, 26.7, and 27.9 eV could be involved in the ΔE for the system. The data are not sufficient to allow a determination as to which process is involved if, indeed, any of the above-mentioned states of nitrogen are involved.

The experimental uncertainty in σ_{10} is ±9.0% and was arrived at by considering the sources and the estimated uncertainties listed in Table I.

Other potential sources of error, i.e., state of ion beam, residual gas scattering, capture in accelerator tube, and target gas impurities, have been reduced to <1% by either experimental arrangement or data acquisition and reduction procedures.

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