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Translational spectroscopy of N^+ produced by collision induced dissociation of N_2^+ on He and Ar

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Abstract

The translational energy is used to suggest the important transitions involved in the production of N^+ in N_2^+ colliding with He and Ar at 3.0 and 4.0 keV. From the energy spectra, interesting structures can be observed. The laboratory energy (LE) distribution of N^+ was transformed to the center of mass (c.m.) assuming the N^+ is isotropically emitted in the c.m. frame. For both cases, the most probable energy was observed with nearly zero kinetic energy released (0.32 ± 0.04 eV for the He target and 0.39 ± 0.05 eV for the Ar target). The transition was associated with the electronic excitation of the C $^2\Sigma_u^+$ state, which predissociates in the continuum of the $^2\Pi_u$ and $^2\Sigma_u$ states, in agreement with the recent results obtained by electron impact dissociation and with a new device which combines imaging and time-of-flight resolved coincidence techniques. It is seen that the N^+ energy distribution for Ar has a wide secondary maxima, while for He, it is either slight or absent. This result is attributed to the dissociation of the D $^2\Pi_g$ state.

Keywords: N_2^+; Translational energy spectroscopy; Collision induced dissociation; Fragmentation mechanism; Ion fragmentation energies

1. Introduction

The distribution of elements in space depends on the processes involved, such as charge transfer, radiative combination, dissociation, electron impact ionization and photoionization [1]. The interstellar space is formed of nitrogen-rich compounds among others. The ionospheres of the Earth and Titan, which have dense atmospheres, are mainly composed of molecular nitrogen. Collisions between N_2 and other gases are likely taking place; it is, therefore, important to study them, in particular at low energies.

N^+ formation has been studied through a variety of low energy experiments like electron capture of N_2 molecules [2], and by N^{2+} [3], fragmentation dynamics [4], charge transfer rate coefficients between N^{2+} and several targets [1,5], electron impact dissociation and ionization of N^{2+} [6], etc.

Most of these experiments were carried out in the energy range in the order of tenths of eV. In this work, we report energy distributions of N^+ from the reactions N_2^+ in He and Ar at the collision energies of 3.0 and 4.0 keV.

2. Experimental approach

Our instrument has been described in detail previously [7]. A schematic diagram of the apparatus is shown in Fig. 1. Briefly, the N_2^+ ions are formed in an arc discharge source, extracted and accelerated to the desired energy. The selected molecular ion beam was velocity analyzed by a Wien filter, passed through cylindrical plates (to deviate it by 10°), and through a series of collimators before entering the gas target cell. This was a cylinder of 2.5 cm in length and diameter, with a 1-mm entrance aperture, and 2-mm-wide, 6-mm-long exit aperture. All apertures and slits had knife edges.

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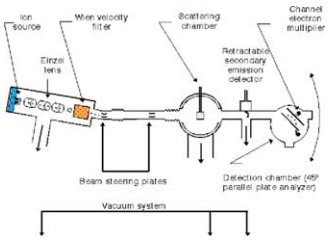

Fig. 1. Schematic diagram of the apparatus.

The target cell, located at the center of a rotatable computer-controlled vacuum chamber that moved the whole detector assembly, is 47 cm away from the detection area. The detector assembly consisted of a Harrower-type parallel-plate analyzer, with a channel electron multiplier (CEM) attached to its exit end. The beam entered the uniform electric field at an angle of 45°. To measure the energy distribution, a 0.36-mm-diameter pinhole was placed at the entrance of the analyzer and a slit 1-cm-long and 0.01-mm-wide was placed in front of the CEM. Path lengths and apertures gave an overall angular resolution of the system of 0.1°.

The energy distributions were obtained by varying the voltage of the analyzer, its calculated energy resolution being $\Delta E/E = 4.5 \times 10^{-3}$. Previous work has shown that dissociation processes exhibit strong dependence upon the ion source condition [8]. Consequently, great care was taken in always having the same ions source parameters.

3. Results and discussion

For the energy distribution it is shown in [9] that the laboratory energy of N^+ can be expressed by:

$$(M_1 + M_2)U = M_1(V_o - W - E) + M_2 W \pm 2[M_1 M_2 W(V_o - W - E)]^{1/2},$$

where U is the final energy of the fragment of mass M_1 detected at an angle of 0° in the laboratory frame, V_o is the initial energy of the molecular projectile of mass $(M_1 + M_2)$, E is the internal energy increase of the system, and W is the kinetic energy released as a result of the dissociation process. The (±) signs correspond to forward or backward ejection of the fragment of mass M_1. Typical laboratory energy spectra for 3.0 and 4.0 keV are shown in Figs. 2 and 3, respectively. The spectra of dissociation energies in collision induced dissociation rarely consist of sharp lines, and the excitation cross sections depend strongly on the instantaneous separation R_{M1M2} of the molecular constituents M_1 and M_2 at the time of collision. Very often, the spectra of dissociation energies consist of broad structures or shoulders due to the distribution of vibrational energies with which the projectile reaches the collision.

In the following analysis, the translational energy is used to suggest the most important transitions involved in the production of N^+ in N_2^+ colliding with He and Ar at collision energies (V_o) of 3.0 and 4.0 keV.

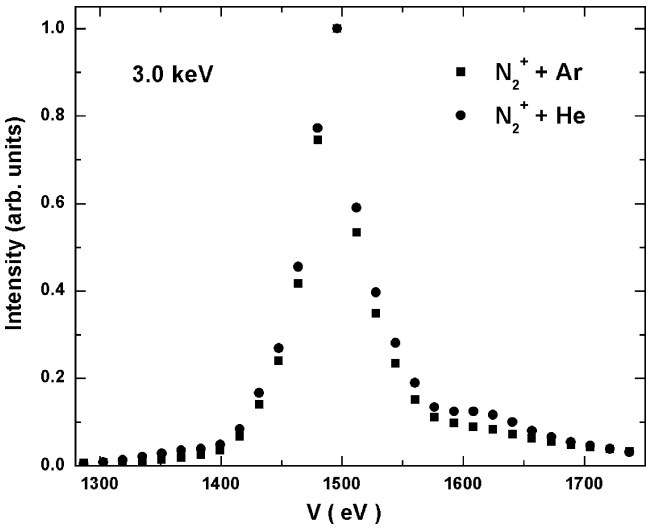

Fig. 2. Laboratory energy spectrum of N^+ resulting from 3.0 keV N_2^+ collisions with He and Ar.

Interesting structures can be observed in the energy spectra. The fine sized detector at 0° is most efficient in collecting N^+ that have zero or near-zero energy in the center of mass (c.m.) of the moving N_2^+. Thus, the single central peak in the energy spectra results from N^+ that dissociate from N_2^+ states with zero energy released ($W \approx 0$). A careful inspection of Figs. 2 and 3 reveals that the concurrent apexes of both energy distributions are slightly shifted to the left of the position that corresponds to zero energy loss, namely, half of the incident energy ($V_o/2$). This shift from the peak at $W = 0$ arises from the loss of energy during the collision, which in turn is a measure of the increment in internal energy of the molecule (E). From the calibration of the analyzer, and from the spectra, a value of $E = 8.5 \pm 0.8$ eV for the energy absorbed by the molecule was determined. This value is in agreement with the recent results obtained by electron impact

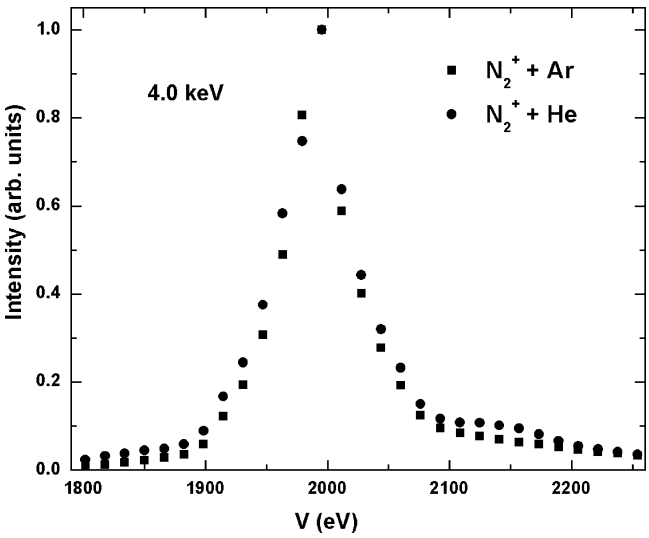

Fig. 3. Laboratory energy spectrum of N^+ resulting from 4.0 keV N_2^+ collisions with He and Ar.

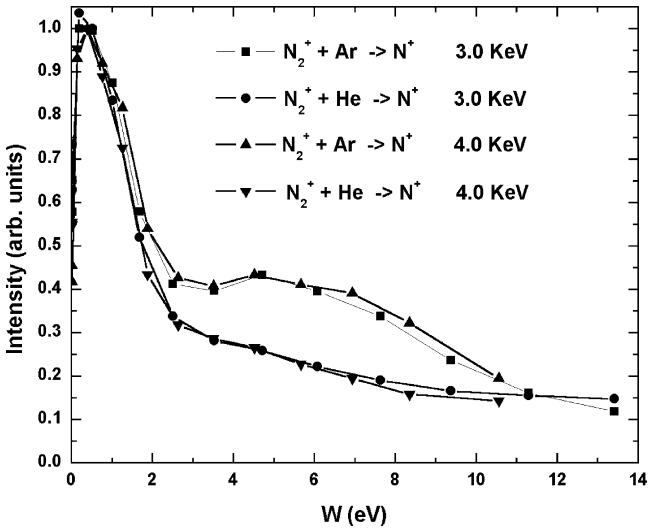

Fig. 4. Energy distribution of N^+ in c.m. of $N_2{}^+$.

dissociation of Bahati et al. [6] and with a new device which combines imaging and time-of-flight resolved coincidence techniques [10]. Consequently, it is reasonable to interpret that the central peak is due to an electronic singlet excitation of the ground state of $N_2{}^+$ to products also formed in their respective ground states ($N(^4S^0)$ and $N^+(^3P)$), meaning that in the threshold region dissociative excitation (DE) takes place via predissociation, as Bahati et al. [6] suggested.

We have transformed the laboratory energy (LE) distribution of N^+ to the center of mass (c.m.) assuming the ion is isotropically emitted in the c.m. frame. The resulting c.m. distributions for $N_2{}^+$ at 3.0 and 4.0 keV for both targets are shown in Fig. 4. For the case of He as a target, the most probable energy was observed at 0.32 ± 0.04 eV. The transition was associated with the electronic excitation of the C $^2\Sigma_u{}^+$ state, which predissociates in the continuum of the $^2\Pi_u$ and $^2\Sigma_u$ states, in agreement with the results obtained by Bahati et al. [6] and with Lafosse et al. [10]. With Ar as a target, the most probable energy liberated by the $N_2{}^+$ was observed at 0.39 ± 0.05 eV. As N^+ ions are produced with nearly zero kinetic energy released, the transitions are attributed to the same processes discussed before for the He case.

The N^+ energy distributions for Ar have a wide secondary maximum (between 4 and 11 eV); while for He, that it is either slight or absent. This result shows that there should be some ionic potentials at 13.4 eV which connect with the dissociation limit of $N(^2D) + N^+(^3P)$ (11.1 eV from the ground sate of the $N_2{}^+$); in this case, the kinetic energy of the N + N^+ is 2.3 eV. For the D $^2\Pi_g$ state, the dissociation limit for which $N(^4S) + N^+(^3P)$ (8.71 eV from the ground sate of the molecule $N_2{}^+$) is the lowest, has been considered to have its Franck–Condon region at about 13.41 eV [11]. If the D $^2\Pi_g$ state dissociates at 13.41 eV, the total kinetic energy released by the fragments is 4.7 eV, which is observed in this experiment for the case of the Ar target. The shape of the energy peak reflects the distribution of translational energy released, and the wide skirts shown by these peaks indicate that a considerable range of energies is involved. Thus, different electronic states and vibrational levels appear to be involved in the experiments. The fact that the energy distributions have different shapes suggests that N^+ arises from different excitation energy levels, and it also reveals the influence of the target. It is interesting to note that the interpretation is based on electronic excitation phenomena.

Since the ion source used in this work is of electron bombardment ionization, the vibrational distribution of the $N_2{}^+$ ions is somewhat uncertain. Henri et al. [12] have estimated this distribution using the appropriate Franck–Condon factors and the electronic state distribution computed by Moran and Friedman [13]. Henri et al. estimated a vibrational distribution for $N_2{}^+(\nu)$ prepared by electron bombardment as 77% for $\nu = 0$; 14% for $\nu = 1$; 5% for $\nu = 2$; and 4% for higher levels. We have assumed that the $N_2{}^+$ ions produced in our source have this same distribution.

We derived the branching ratio from the ratio of the areas for the N^+ production (Figs. 2 and 3) by $N_2{}^+$ on He to that for $N_2{}^+$ on Ar, at 3.0 and 4.0 keV. From this derivation, we found $\sigma_{Ar}/\sigma_{He} = 1.26$ and 1.53 for the collision energies of 3.0 and 4.0 keV, respectively. We can infer that the cross section for the production of N^+ is a factor of 1.4 times larger for the $N_2{}^+$ + He system than for the $N_2{}^+$ + Ar system.

In conclusion, we have shown the importance of the target in the collisions studied. Despite the fact that the principle maximum in the c.m. distributions for $N_2{}^+$ at 3.0 and 4.0 keV are close for the targets considered, a secondary maximum is clearly evident only for the Ar target. Also, we have proposed the molecular states involved in the dissociation processes studied. It would be interesting to investigate the behavior of $N_2{}^+$ with the other rare gases as targets, and determine the dependence on Z, on the incident energy and on the cross section for the N^+ production.

Acknowledgments

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