Kinetic-energy release in N₂ fragmentation by charge-changing collisions of 2-MeV C⁺ ions

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Collision-induced fragmentation of N₂ was investigated for 2-MeV C⁺ ions under charge-changing conditions of $C^+ \rightarrow C^{q+}$ ($q = 0, 2, 3$). Coincidence measurement of fragment ions was performed by means of a momentum three-dimensional imaging technique at scattering angles of $\theta = 0$ and 1.0 mrad. Kinetic-energy release (KER) obtained for a fragmentation channel of $N_2^{2+} \rightarrow N^+ + N^+$ was found to differ significantly in electron loss and capture collisions. In two-electron-loss collisions ($C^+ \rightarrow C^{3+}$), KER spectra were essentially identical for $\theta = 0$ and 1.0 mrad. It is concluded that the energy level of dissociating excited states of N_2^{2+} may be saturated when the interaction strength, defined as $\frac{q}{vb}$, exceeds 0.65, where q and v are the charge and the velocity, respectively, of an incident ion, and b is the impact parameter.

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Molecular fragmentation and ionization have been extensively studied so far by using various projectile ions [1]. In recent investigations three-dimensional momentum imaging techniques were used to determine molecular orientation [2–6] and kinetic-energy release (KER) during fragmentation [7–13]. As the molecular fragmentation occurs predominantly from a dissociative excited state, it is important to measure KER in individual fragmentation channels because it carries direct information about the initial dissociative excited states [8,14]. The amount of electronic energy deposition into a target molecule is the essential quantity that characterizes the fragmentation of molecules ranging from diatomic [3] to polyatomic [15] and cluster particles [16].

The amount of energy deposition may be estimated if the impact parameter (b) between collision partners is known. For fast heavy ions, however, b-resolved measurements have rarely been performed so far except those of cold target recoil-ion momentum spectroscopy [1] and those performed under projectile charge-changing conditions [6,11–13,17–21]. In slow collisions of He^{2+} and Ar^{3+} with N₂, Ehrich *et al.* [11] found that KER spectra of two fragment N⁺ ions are largely different for individual projectile final charge states.

In our recent study of CO fragmentation in charge-changing collisions of B^{2+}, O^{2+}, and Si^{2+} ions [12], the fragmentation was found to differ strongly depending on the type of charge changing and on the projectile species. The former result means that the effective impact parameters are different in electron-loss and -capture collisions, leading to different energy deposition in these collisions. The latter implies that the magnitude of the binding energy of the relevant electronic states of a projectile ion play a role in both loss and capture collisions.

In order to achieve more precise understanding of impact-parameter-dependent molecular fragmentation, we performed a scattering-angle (θ) resolved coincidence experiment for charge-changing collisions between 2-MeV C⁺ and N₂. The experiment was done at $\theta = 0$ and 1.0 mrad. Time-of-flight

mass spectra and KER distributions in a fragmentation channel of $N_2^{2+} \rightarrow N^+ + N^+$ are presented for one-electron- (1*e*-) capture, 1*e*-loss, and 2*e*-loss collisions. Experimental results are discussed using the interaction strength $\frac{q}{vb}$ [21], where q and v are the projectile charge and velocity, respectively, and b is the impact parameter.

The experiment was carried out at the Quantum Science and Engineering Center 1.7-MV tandem accelerator facility of Kyoto University. The experimental method and apparatus are described elsewhere [6,12,17], so only a brief outline is given below. A beam of 2-MeV C⁺ was carefully collimated ($\leqslant 0.5$ mm in diameter) and was incident on an effusive molecular beam target of N₂. A base pressure of a collision chamber was kept below 1×10^{-5} Pa. After collisions, final charge states of projectile particles were separated by an electrostatic beam deflector and detected by a movable solid-state detector (SSD). We installed a hole slit of 0.5 mm diam in front of the SSD to resolve the scattering angle θ. An acceptance angle of the detector was 0.5 mrad. Fragment ions as well as intact ions produced in collisions were extracted by an electric field of 423 V/cm and detected by a position- and time-sensitive delay line detector. Mass spectra were obtained by a time-of-flight (TOF) method under a Wiley-Mclaren spatial focusing condition. Extraction of fragment ions into a TOF spectrometer was achieved over 4π sr for all fragment ions investigated. A total detection efficiency, given by the mesh-transmission rate (0.613) multiplied by the multichannel plate open area (0.57), was 0.35 [12]. The event-by-event recording was made with a digital storage oscilloscope (Wavepro7000, LeCroy), enabling us to measure flight times t and positions (x, y) of plural fragment ions produced in a single event. The amounts of KER of fragments were obtained from t and (x, y) with an energy resolution of about 1 eV.

Figure 1 shows TOF spectra obtained for 1*e*-capture, 1*e*-loss, and 2*e*-loss collisions of 2-MeV ($v = 2.58$ a.u.) C⁺ ions at scattering angles of $\theta = 0 \pm 0.5$ and 1.0 ± 0.5 mrad. The spectra show fragment ions of N^{k+} ($k = 1$–3) and ionized molecular ions of N_2^+ and N_2^{2+}. The width of each spectrum reflects the magnitude of the kinetic energy of corresponding ions. It is obvious that the fragment ions have kinetic energies much larger than those of intact parent ions. Note a sharp line of N_2^{2+} superimposed on a broad spectrum of

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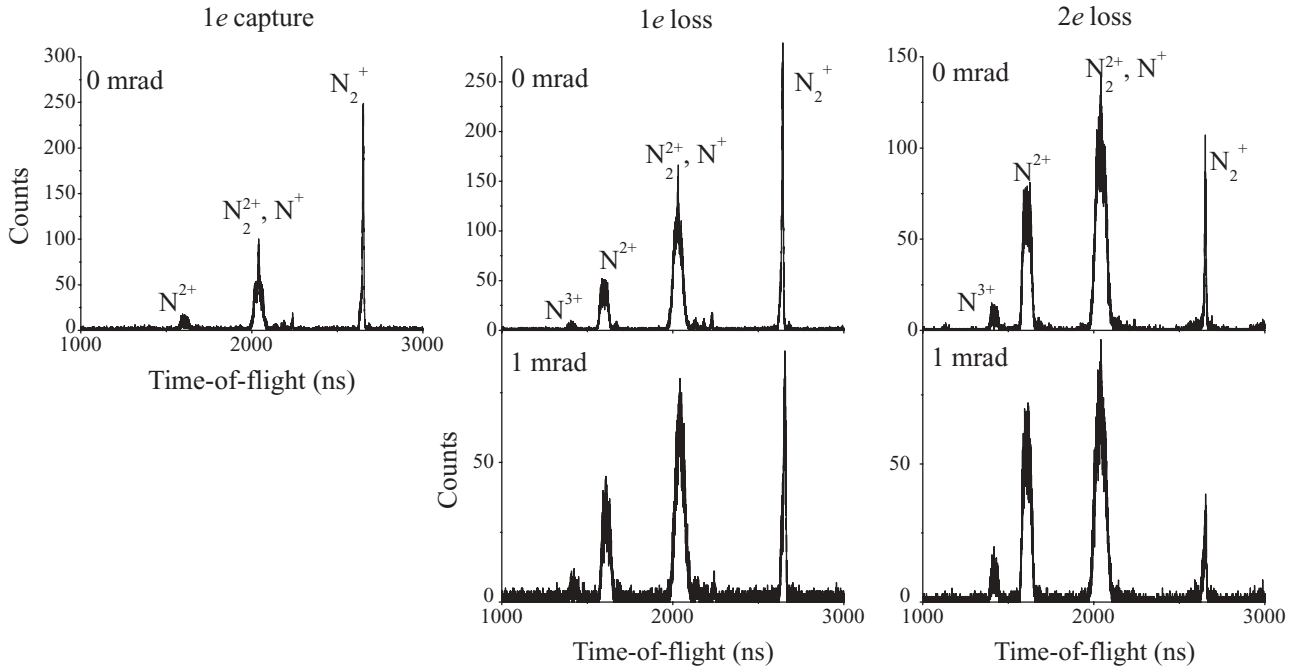
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FIG. 1. Time-of-flight spectra obtained in charge-changing collisions of 2 MeV C$^+$ ions at $\theta = 0$ and 1.0 mrad.

N$^+$. In 1e-capture collisions, almost no peaks were observed at $\theta = 1.0$ mrad, indicating that single-electron capture occurs in distant collisions.

By contrast, in electron-loss collisions, particularly in 2e loss, peak intensities of fragment ions are considerably larger than singly charged parent ions N$_2^+$. These features are nearly the same as for CO fragmentation studied by us using B^{2+}, O^{2+}, and Si^{2+} ions at an energy of 71.4 keV/nucleon ($v = 1.69$ a.u.) [12]. As pointed out in [6,12], ionization (electron loss) of C$^+$ by a neutral molecule may be limited to only small impact-parameter collisions. This is because a somewhat large inelastic energy is required to remove a tightly bound projectile electron [i.e., $I($C$^+) = 24.38$ eV and $I($C$^{2+}) = 47.89$ eV]. Such a close collision may easily cause multiple ionization and fragmentation. Scattering-angle dependence in electron-loss collisions shows that the intensity of fragment ions N^{k+} relative to the parent ion N$_2^+$ increases at $\theta = 1.0$ mrad in comparison with 0 mrad. As the degree of fragmentation depends on the amount of inelastic energy transferred to a target molecule, the present result simply means that smaller b collisions accompany larger energy deposition. Note that the impact parameter corresponding to $\theta = 1.0$ mrad is estimated to be about 0.6 a.u. for the collision of 2-MeV C with N.

Figure 2 shows KER spectra measured at $\theta = 0$ mrad in a fragmentation channel of N$_2^{2+} \rightarrow$ N$^+$ + N$^+$ for individual charge-changing processes. In 2e-loss collisions, the KER spectra show a broad distribution ranging from 10 to 30 eV, while in other collisions peak intensities at higher than 15 eV are about half that of 2e-loss collisions. In particular, the KER in 1e capture is narrow and exhibits a few prominent peaks at about 10 and 17 eV. It implies obviously that 2e-loss collision populates higher excited states of N$_2^{2+}$, which is consistent with our previous results obtained for CO [12]. In electron impact experiments, Lundqvist *et al.* [14] found three dissociative

states of N$_2^{2+}$ with characteristic energy releases of about 8, 10, and 15 eV. The peak at 10 eV is assigned to the $D\ ^1\Sigma_u^+$ electronic state.

Figure 3 shows the scattering-angle dependence of KER in 1e- and 2e-loss collisions at $\theta = 0$ and 1.0 mrad. In 1e-loss collisions, the KER spectrum at $\theta = 1.0$ mrad is slightly higher than that at $\theta = 0$ mrad, while in 2e-loss collisions the KER spectra are found to be essentially the same for both angles. It implies that similar excited states of the molecular ion are populated at both scattering angles.

A similar remark is reported in fast heavy-ion collisions with N$_2$ [9], stating that the KER spectra are almost identical for 5.9 MeV/amu xenon ions and 4.7 MeV/amu bismuth ions. They examined the results in terms of the Sommerfeld parameter $\kappa = \frac{q}{v}$ and concluded that the shape and width
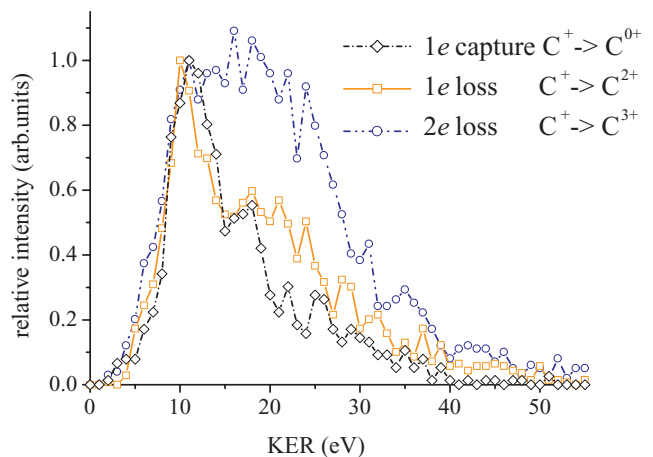

FIG. 2. (Color online) N$^+$ + N$^+$ KER spectra obtained in charge-changing collisions of 2-MeV C$^+$ at $\theta = 0$ mrad.
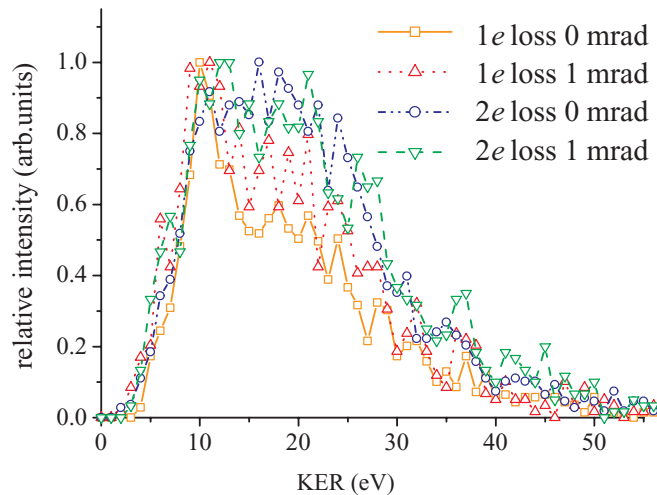
FIG. 3. (Color online) $N^+ + N^+$ KER spectra obtained in charge-changing collisions of 2-MeV C^+ at $\theta = 0$ and 1.0 mrad.

of KER spectra become saturated for strong interaction of $\kappa > 1$. In the present experiment, however, the Sommerfeld parameter κ is only 0.39 and the interaction is not strong within this model. Hence, our experimental results suggest that the κ parameter cannot be applied straightforwardly to low-q collisions. More important, the Sommerfeld parameter is not appropriate anyway because it does not take b into consideration. Instead of the κ parameter, we used the interaction strength $\frac{q}{vb}$, which is more useful to characterize the molecular fragmentation as demonstrated in [21]. In our case, it reads $\frac{q}{vb} = 0.65$ for $b = 0.6$ a.u. ($\theta = 1.0$ mrad).

Consequently, we conclude that the KER becomes saturated when the interaction strength is larger than 0.65 at least.

On the other hand, the mass spectra shown in Fig. 1 exhibit high enhancement of fragment ions at $\theta = 1.0$ mrad, as described above. Hence, we interpret these results in the following way. Namely, a N_2 molecule is at first populated to highly excited states at smaller b collisions and it may decay promptly to lower dissociating excited states via autoionization [22]. Consequently, the energy level of dissociating excited states of N_2^{2+} becomes saturated with increasing interaction strength, although the total energy deposition may increase [3,4].

In summary, we studied the fragmentation of N_2 in charge-changing collisions of 2-MeV C^+ ions at $\theta = 0$ and 1.0 mrad. It was found that multiple ionization and fragmentation are enhanced in electron-loss collisions compared to capture collisions and more prominently at large scattering angles. The different features between loss and capture collisions may be attributed to the difference of effective impact parameters associated with the type of charge-changing collisions. The KER spectra for the fragmentation channel $N_2^{2+} \rightarrow N^+ + N^+$ are also different, implying that excited molecular states are different according to the type of charge changing. We also found that, in 2e-loss collisions at $\theta = 0$ and 1.0 mrad, the KER spectra are nearly the same, while the mass spectra exhibit larger fragmentation at $\theta = 1.0$ mrad. It implies that, in 2e-loss collisions, the dissociating excited states formed after relaxation via autoionization may be saturated, whereas the fragmentation as well as multiple ionization is enhanced with increasing amounts of energy deposition into a target molecule. The present results suggest that such saturation appears to occur at the interaction strength $\frac{q}{vb}$ of 0.65. At present we are planning to examine this saturation effect in more detail by changing the initial charge state of incident carbon ions.

[1] J. Ullrich, R. Moshammer, A. Dorn, R. Döner Schmidt, and H. Schmidt-Böcking, Rep. Prog. Phys. **66**, 1463 (2003).

[2] U. Werner, N. M. Kabachnik, V. N. Kondratyev, and H. O. Lutz, Phys. Rev. Lett. **79**, 1662 (1997).

[3] B. Siegmann, U. Werner, Z. Kaliman, Z. Roller-Lutz, N. M. Kabachnik, and H. O. Lutz, Phys. Rev. A **66**, 052701 (2002).

[4] B. Siegmann, U. Werner, H. Lebius, B. Huber, H. O. Lutz, and R. Mann, Nucl. Instrum. Methods B **205**, 629 (2003).

[5] I. Ali et al., Nucl. Instrum. Methods B **149**, 490 (1999).

[6] T. Mizuno, T. Majima, H. Tsuchida, Y. Nakai, and A. Itoh, J. Phys.: Conf. Ser. **58**, 173 (2007).

[7] L. Adoui, C. Caraby, A. Cassimi, D. Lelièvre, J. P. Grandin, and A. Dubois, J. Phys. B **32**, 631 (1999).

[8] M. Tarisien et al., J. Phys. B **33**, L11 (2000).

[9] B. Siegmann, U. Werner, R. Mann, N. M. Kabachnik, and H. O. Lutz, Phys. Rev. A **62**, 022718 (2000).

[10] J. Rajput, S. De, A. Roy, and C. P. Safvan, Phys. Rev. A **74**, 032701 (2006).

[11] M. Ehrich, B. Siegmann, U. Werner, and H. Lebius, Radiat. Phys. Chem. **68**, 127 (2003).

[12] T. Mizuno, T. Yamada, H. Tsuchida, Y. Nakai, and A. Itoh, Phys. Rev. A **81**, 012704 (2010).

[13] S. De, P. N. Ghosh, A. Roy, and C. P. Safvan, Nucl. Instrum. Methods B **243**, 435 (2006).

[14] M. Lundqvist, P. Baltzer, D. Edvardsson, L. Karlsson, and B. Wannberg, Phys. Rev. Lett. **75**, 1058 (1995).

[15] N. Neumann et al., Phys. Rev. Lett. **104**, 103201 (2010).

[16] H. Tsuchida, A. Itoh, K. Miyabe, Y. Bitoh, and N. Imanishi, J. Phys. B **32**, 5289 (1999).

[17] T. Mizuno, T. Yamada, H. Tsuchida, Y. Nakai, and A. Itoh, J. Phys.: Conf. Ser. **163**, 012039 (2009).

[18] R. L. Watson, G. Sampoll, V. Horvat, and O. Heber, Phys. Rev. A **53**, 1187 (1996).

[19] T. Kaneyasu, T. Azuma, and K. Okuno, J. Phys. B **38**, 1341 (2005).

[20] S. Tsurubuchi and K. Motohashi, J. Phys. B **36**, 3847 (2003).

[21] E. Wells, T. Nishide, H. Tawara, R. L. Watson, K. D. Carnes, and I. Ben-Itzhak, Phys. Rev. A **77**, 064701 (2008).

[22] E. Shigemasa, T. Kaneyasu, T. Matsushita, Y. Tamenori, and Y. Hikosaka, New J. Phys. **12**, 063030 (2010).