The role of nuclear stopping power in fast heavy ion collision with C_{60}

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Abstract

Velocity dependence of C_{60} fragmentation induced by neutral Au^0 projectiles ($v = 0.2$–1.1 a.u.) is studied. It is observed that the fragmentation patterns depend strongly on the collision velocity, and the velocity dependence is strikingly different from recent experiments for H^+ and He^+ ions measured in the same velocity range. This is attributed to a largely different total amount of excitation energy induced by Au^0 and light ions. We also found the multifragmentation pattern to change dramatically as a function of the projectile velocity, indicating obviously that the nature of excitation (vibrational or electronic) affects the fragmentation mechanism. Furthermore, intensity ratios between small fragment ions and C_{60}^+ fall to a minimum at $v \sim 0.5$ a.u., showing a clear evidence of "transparency window" where vibrational and electronic excitation are both weak. © 2002 Elsevier Science B.V. All rights reserved.

1. Introduction

Study of collision dynamics between an energetic ion and a C_{60} molecule leads to understand ion–solid interactions from a microscopic viewpoint, because C_{60} molecule is one of the most ideal atomic many-body systems. A lot of experimental and theoretical studies using fullerenes as collision partners have been performed in the last decade and the collision is called "fullerene collision" or "fullerene reaction" [1,2]. One of the most exciting discussion in fullerene collisions is how the excitation mechanism, i.e. vibrational or electronic, affects relaxation and fragmentation of an excited C_{60} after collisions [3–6].

Up to now, two different fragmentation mechanisms have been reported experimentally for different projectiles (H^+ and $He^{1,2+}$) [3–5]. In He ion collisions at $v = 0.1$–1 a.u., a transition from evaporation to multifragmentation was observed to occur at $v \sim 0.5$ a.u. [3,4]. The authors concluded that the vibrational excitation leads to evaporation and the electronic excitation triggers multifragmentation. On the other hand, contradicting results were observed in H^+ collisions at $v = 0.2$–3.5 a.u. That is, velocity dependence of evaporation fractions is similar to that of the electronic stopping power rather than the nuclear

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stopping power [5]. It shows that the electronic excitation has an important role in evaporation processes.

Recently, a microscopic calculation based on the non-adiabatic quantum molecular dynamics theory (NA-QMD) was applied to collisions between C_{60} and H^+, C^+, Ar^+ ions ($v = 0.01$–0.5 a.u.) [6]. The calculations show a drastic change of fragmentation dynamics with increasing collision velocity, suggesting that the fragmentation pattern is affected by the difference of excitation mechanisms.

In this work, we performed collision experiments using neutral Au^0 projectiles in the velocity range from 0.2 a.u. (0.2 MeV) to 1.1 a.u. (6.0 MeV). Nuclear energy deposition is supposed to be considerably higher than those for H or He ions due to a large nuclear charge. In this velocity region, a transition from vibrational to electronic excitation is expected, as predicted both in the stopping power consideration and in the microscopic calculations [6]. Furthermore, in neutral–neutral collisions, energy transfer in distant collisions is strongly reduced, since projectile atoms have to penetrate or graze an electronic cloud of C_{60} in order to cause Coulomb interactions. In distant collisions, electronic excitation is considered to be predominant. As a result, the relative importance of nuclear energy transfer is expected to be strongly enhanced because of absence of distant collisions.

2. Experiment

The experiment was performed at the 1.7 MV tandem Cockcroft–Walton accelerator facility of Kyoto University. A beam of Au ions from the accelerator was collimated to about 1×1 mm^2 with two sets of four-jaw slits. A neutral beam was produced via electron-capture collisions with residual gases in the beam line. Then, the primary ion beam was removed by a magnet. A C_{60} molecular target was obtained by sublimation of pure C_{60} powder at 450 °C in a quartz oven. Mass to charge distributions of produced ions were measured by a time-of-flight (TOF) method under a Wiley–McLaren spatial-focusing condition. The

detailed TOF techniques are described in our previous papers [8,9]. In this experiment, secondary electrons emitted in collisions are used as trigger signals of the TOF measurements. The base pressure was below 1×10^{-5} Pa through the whole experiment.

3. Results and discussion

Fig. 1 shows TOF spectra obtained for collisions of Au^0 with C_{60} at different collision energies. It is seen that small fragment ions C_n^+ ($n < 15$), produced via multifragmentation are the predominant products over the whole energy range investigated. It indicates that a large amount of energy is transferred into C_{60} in Au^0 collisions. Tsuchida et al. estimated the internal excitation energy of 794 eV for Au^{3+} ions ($v = 0.9$ a.u.) from
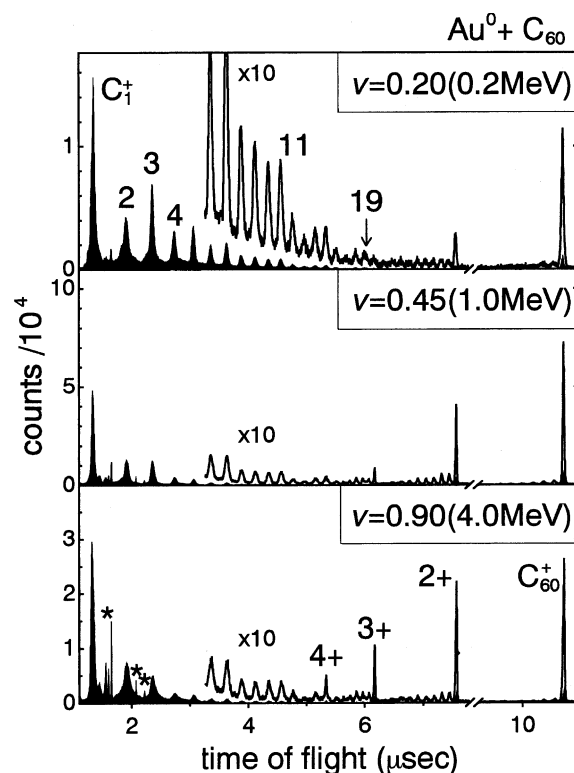

Fig. 1. TOF spectra obtained in collisions of Au^0 with a C_{60} molecule at different velocities. The peaks shown by (*) originate from background residual gases.
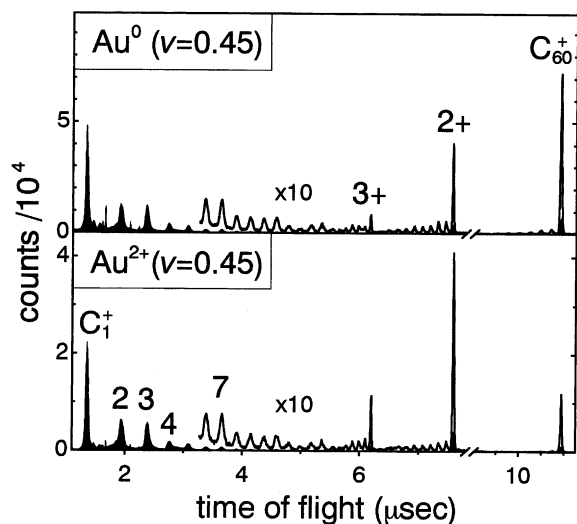
Fig. 2. TOF spectra obtained in collisions of Au^0 and Au^{2+} with a C_{60} molecule at the same velocities ($v = 0.45$ a.u.).

charge state distributions are probably attained [8,9].

Projectile velocity dependence of relative intensities of product ions with respect to the C_{60}^+ intensity is plotted in Fig. 3. Intensities of intact ions of C_{60}^{r+} ($r = 2, 3$) increase monotonously with increasing velocity. Since a degree of ionization is directly related to the electronic excitation, it is confirmed that the electronic excitation energy increases monotonously in this velocity region as predicted in stopping power considerations.

The most striking feature of the present results is a U-shaped velocity dependence observed for small fragment ions C_n^+ ($n < 15$) in Fig. 3. The relative intensity changes dramatically around $v = 0.45$ a.u. The U-shaped dependence suggests following two nature of multifragmentation,
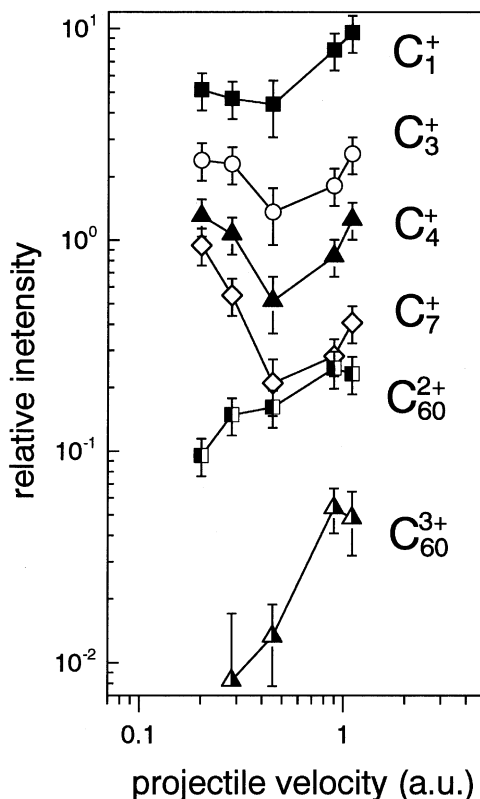
a stopping power consideration [7]. This energy is considerably higher than a calculated value of 225 eV required to induce entire multifragmentation [10].

Fig. 2 shows a comparison of TOF spectra for Au^0 and Au^{2+} projectiles at the same velocity ($v = 0.45$ a.u.). Both spectra were measured in coincidence with secondary electrons as mentioned above. It is seen that the peak intensities of C_{60}^+, C_{60-2m}^+ are strongly suppressed in Au^{2+} collisions due to pure single electron-capture collisions where no free electrons are emitted. Although the production of C_{60}^{2+} is also supposed to be suppressed in Au^{2+} collisions due to double electron capture, the relative intensity of C_{60}^{2+} with respect to the C_1^+ intensity is higher for Au^{2+} than Au^0 projectiles. Since the most significant difference between Au^{2+} and Au^0 projectiles appears in distant collisions, the above results indicate that C_{60}^{2+} are produced mainly in distant collisions for Au^{2+} projectiles, and probably in grazing collisions for Au^0 projectiles. On the other hand, distributions of small fragment ions are close to each other. The charge-independent distribution implies that the multifragmentation occurs in penetrating collisions since initial charge states of projectiles may be smeared inside a C_{60} cage and quasiequilibrium

Fig. 3. Velocity dependence of relative intensities of multiply charged intact ions C_{60}^{r+} ($r = 3$ and 2) and small fragment ions C_n^+ ($n = 1, 3, 4, 7$) with respect to the C_{60}^+ intensity.

(1) the vibrational excitation plays also an important role in multifragmentation, as well as the electronic excitation, for Au^0 projectiles, (2) different multifragmentation patterns are led via vibrational and electronic excitation. First, rapid decreases of relative intensities below $v < 0.45$ a.u. cannot be explained by only a contribution of the electronic excitation, which increases monotonously as described above. Instead, it indicates that the vibrational excitation plays an important role in Au^0 collisions. This is completely different from the results obtained in He ion collisions where the relative intensities of small fragment ions C_n^+ ($n < 15$) increase monotonously and the vibrational excitation less affects the multifragmentation [3,4]. Schlatholter et al. estimated the projectile energy losses of He ions using molecular dynamics simulations. According to their calculations, elastic collisions transfer an energy of only few eV in He collisions at $v = 0.27$ a.u. This energy is comparable or less than the dissociation energy of 10 eV [11], and cannot lead to multifragmentation which needs at least more than 80 eV internal energy [10]. In Au collisions, a large amount of energy is supposed to be deposited via elastic collisions. From the comparison between Au^0 and He^+ projectiles, it is concluded that a total amount of energy is decisive to multifragmentation. A similar projectile dependence of fragmentation patterns has been reported for collisions of $C_{60}^{\pm}$ with He, Ne, Ar atoms in lower collision velocity region ($v \sim 0.05$ a.u.) where the vibrational excitation is predominant [12]. Relative intensities of small fragmentation ions were predominant in collisions with Ar. On the contrary, few small fragment ions were observed in He collisions.

Second, one can see medium size fragment ions C_n^+ ($n = 6$–20) in Fig. 3 to decrease steeply in $v < 0.45$ a.u. (vibrational domain) and increase more slowly than smaller fragment ions in $v > 0.45$ a.u. (electronic domain). This effect is also seen in TOF spectra (Fig. 1), where the distribution of C_n^+ ($n < 15$) shows an even–odd oscillation for $v < 0.45$ a.u. and an exponential decay for higher velocities. Namely, the electronic excitation enhances multifragmentation leading to a catastrophic break-up of a fullerene cage. On the contrary, multifragmentation via vibrational exci-

tation appears to contribute to the production of medium size fragment ions.

From another viewpoint, the U-shaped dependence is a clear evidence of "transparency window" where vibrational and electronic excitation are both weak, as predicted theoretically for Na_9^+–He collisions [13] and in He^+–C_{60} experiments where a transition from evaporation to multifragmentation was observed at $v \sim 0.5$ a.u. [3].

Finally, we discuss velocity dependence of evaporation fractions f_e^{r+} defined by

$$f_e^{r+} = \sum_{m=1}^{m_{\max}} I(C_{60\text{-}2m}^{r+}) \bigg/ \sum_{m=0}^{m_{\max}} I(C_{60\text{-}2m}^{r+}),$$

where $I(C_{60\text{-}2m}^{r+})$ represents a peak integral of $C_{60\text{-}2m}^{r+}$ and $m_{\max} = 3$ for $r = 1$ and 6 for $r = 2$. Fig. 4(a) shows f_e^{r+} obtained in the present experiments. Results obtained in other experiments for H^+ and He^+ ions are also plotted in Fig. 4(b) [3,5]. The evaporation fraction for Au^0 decreases monotonously with increasing velocity. The dependence is similar to that for He^+ collisions, but is different from H^+ collisions. At present, there are no theoretical studies to explain the behavior of the ve-
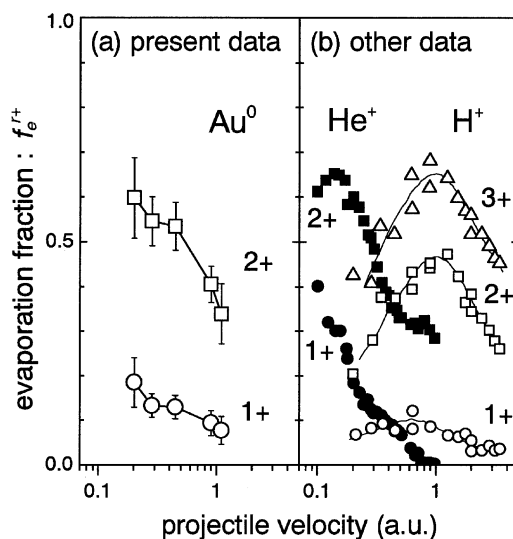

Fig. 4. Velocity dependence of evaporation fractions f_e^{r+} for Au^0, H^+ and He^+ collisions. Data for H^+ and He^+ collisions are taken from [3–5]. The fraction for 2^+ at 0.2 a.u. in Au^0 collisions is probably overestimated due to the superimposition of C_n^+ for $n = 24$–28.

locity dependence for all projectiles. Opitz et al. concluded, from Monte Carlo calculations of the energy loss in H^+ collisions, that the vibrational excitation is still considerably lower than the electronic excitation in this velocity region [5]. Hence, it is considered that the effect of the nuclear energy transfer has less contribution to evaporation in H^+ collisions. It is interesting to note that a total amount of excitation energy seems to play a decisive role in evaporation processes as well as in multifragmentation. Note, however, that a total amount of energy cannot explain the monotonously decreasing behavior observed in He^+ and Au^0 collisions.

4. Conclusions

Velocity dependence of C_{60} fragmentation patterns induced by neutral Au^0 projectiles ($v = 0.2$–1.1 a.u.) was studied. We found that relative intensities of small fragment ions C_n^+ ($n < 15$) with respect to C_{60}^+ reveal a U-shaped velocity dependence. It indicates that the vibrational excitation plays an important role as well as the electronic excitation in multifragmentation by Au^0 projectiles. Thus, it is concluded that a total excitation energy plays an essential role in C_{60}-multifragmentation. It was also found that the multifragmentation pattern changes drastically with increasing velocity, showing that the electronic excitation enhances the degree of multifragmentation, while the vibrational excitation contributes to the production of medium size fragment ions C_n^+ ($n = 6$–20). It implies obviously that the different excitation mechanism (vibrational or electronic) leads to different fragmentation patterns. Furthermore, a minimum relative intensities observed for small fragment ions at $v = 0.45$ a.u. is

certainly a clear experimental evidence of transparency window [13]. The present velocity dependence is found to be essentially different from light projectile ions like H^+ and He^+ [3–5]. This is again attributed to a largely different total excitation energy induced by Au^0 and light ions. Further study is needed to know to what extent the electronic excitation contributes to evaporation and multifragmentation of fullerene molecule.

Acknowledgements

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