Energy loss of fast Li and Si projectiles in electron loss collisions with $C_{60}$

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Abstract

We have studied the multifragmentation of $C_{60}$ under electron loss collisions of 2 MeV Li$^{0,2+}$ and Si$^{0,4+}$ projectiles. Assuming the Bragg’s additivity rule and using available stopping power program codes, electronic energy deposition into a $C_{60}$ molecule has been calculated for six and 15 different electron loss collisions of Li and Si projectiles, respectively. To do this, the impact-parameter dependent ionization probability of incident particles was calculated semiclassically. The calculated energy deposition was found to have a remarkably simple relationship with the degree of $C_{60}$-multifragmentation obtained experimentally for Si projectiles. From comparisons between calculations and experiments, it is also suggested that the stopping power partition rate leading to internal excitation of $C_{60}$ may be significantly different for Li and Si projectiles.

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1. Introduction

Energy loss of energetic particles in matter is the most fundamental collision phenomena studied for many decades [1]. A projectile particle loses its kinetic energy via a variety of interaction mechanisms depending largely on the velocity and the nuclear charge of the incident particle and also on the nature of the target particles. In general, it is well established that the dominant energy loss mechanism is excitation and ionization of the target particles at high velocities exceeding the Bohr velocity $v_0 = 2.2 \times 10^8$ cm/s. For polyatomic molecular targets like $C_{60}$, the deposited energy is spent not only for electronic excitation and ionization but also for vibrational excitation, leading to molecular fragmentation.

To date, considerable effort has been devoted to study of collision-induced $C_{60}$ fragmentation as reported in a recent review article [2] and references therein. Quantitative understanding of the relationship with the projectile energy loss is, however, much less achieved. Energy loss measurements have been carried out only for very low velocity collisions [3–6]. Theoretical investigations are also limited to the velocity region below $v_0$. 
Recently, the Bielefeld group [10] has extended calculations to fast protons of velocities ranging from \(2v_0\) to \(9v_0\) using the local density approximation (LDA) based on the free-electron gas model [11,12]. In terms of the calculated energy deposition, they discussed successfully various experimental results such as \(C_2\)-evaporation rate and ionization cross sections.

The multifragmentation of \(C_{60}\) is also induced rather strongly in charge-changing collisions of fast ions [13–15]. This fact implies clearly that a large amount of inelastic energy is deposited into a \(C_{60}\) molecule in these collisions. On the other hand, it is known that, in fast ion–atom collisions, the amount of projectile energy loss accompanying charge-changing collisions is negligibly small in comparison with that accompanying target ionization [16–18]. To shed some light into this problem, we attempted recently to estimate the amount of energy deposition for 2 MeV Si\(^q^{+}\) (\(q = 0, 1, 2, 4\)) ions undergoing electron loss collisions with \(C_{60}\) [15].

In this work, a comprehensive set of calculations are performed for all the combinations between \(q = 0–4\) and outgoing charge states of \(k = q + 1–5\). The calculation is also extended to the electron loss collisions of light projectile ions of 2 MeV Li\(^q^{+}\) (\(q = 0–2\)). The calculation is carried out for the impact-parameter (\(b\)) dependent ionization probabilities of projectile ions using well-known tables of semiclassical approximation (SCA) calculation given by Hansteen et al. [19] and the \(b\)-dependent stopping function for C atom using the CasP code developed by Grande et al. [20,21]. By combining these two quantities the energy deposition in individual electron loss collisions with \(C_{60}\) is estimated [22–24] including neutral projectiles [14,15]. Consequently, the mass distribution of product ions is composed of two parts corresponding to a series of singly charged small fragment ions of \(C_n^+\) \((n \geq 1)\) and ionized parent/daughter ions of \(C_{60}^{r-m}\) \((m, r \geq 1)\). The former part is created via catastrophic molecular disintegration (multifragmentation) caused by cage-penetrating collisions, and the ionization part is created via relatively soft and larger impact-parameter collisions. The degree of multifragmentation is supposed to depend on the amount of transferred energy from incident particles. Thus, intensity distribution of small fragment ions varies significantly for different projectile species. A typical example of the \(C_n^+\)-intensity variation obtained for various projectile ions is demonstrated in Fig. 1 [24]. The data were obtained by means of time-of-flight (TOF) coincidence techniques between fragment ions and pulsed incident beams, and therefore contain all the contributions from outgoing charge states. It is seen clearly that the intensities of larger sized ions decrease rapidly with increasing projectile atomic number, indicating undoubtedly that a larger amount of energy is transferred from heavier projectiles. The intensity variation as a function of the cluster size \(n\) can be well expressed by a power-law form of \(Y(n) \sim n^{-\lambda}\) for heavier projectile ions. The power \(\lambda\) is supposed to have a close relationship with the amount of energy deposition, and consequently it may reflect fairly well the degree of multifragmentation as discussed in [24].

2. Multifragmentation of \(C_{60}\)

Fragmentation as well as ionization of \(C_{60}\) is commonly induced by MeV energy heavy particles [22–24] including neutral projectiles [14,15]. Consequently, the mass distribution of product ions is composed of two parts corresponding to a series of singly charged small fragment ions of \(C_n^+\) \((n \geq 1)\) and ionized parent/daughter ions of \(C_{60}^{r-m}\) \((m, r \geq 1)\). The former part is created via catastrophic molecular disintegration (multifragmentation) caused by cage-penetrating collisions, and the ionization part is created via relatively soft and larger impact-parameter collisions. The degree of multifragmentation is supposed to depend on the amount of transferred energy from incident particles. Thus, intensity distribution of small fragment ions varies significantly for different projectile species. A typical example of the \(C_n^+\)-intensity variation obtained for various projectile ions is demonstrated in Fig. 1 [24]. The data were obtained by means of time-of-flight (TOF) coincidence techniques between fragment ions and pulsed incident beams, and therefore contain all the contributions from outgoing charge states. It is seen clearly that the intensities of larger sized ions decrease rapidly with increasing projectile atomic number, indicating undoubtedly that a larger amount of energy is transferred from heavier projectiles. The intensity variation as a function of the cluster size \(n\) can be well expressed by a power-law form of \(Y(n) \sim n^{-\lambda}\) for heavier projectile ions. The power \(\lambda\) is supposed to have a close relationship with the amount of energy deposition, and consequently it may reflect fairly well the degree of multifragmentation as discussed in [24].

![Figure 1: Intensity distribution of \(C_n^+\) ions from \(C_{60}\) induced by collisions with various projectile ions [24].](image-url)
Similar multifragmentation spectra are also produced in electron capture and loss collisions \[13–15\]. Fig. 2 shows examples measured for electron loss collisions of 2 MeV Li and Si ions, where the TOF spectra were measured in coincidence without going projectile charge states. Some remarkable differences can be pointed out in the intensity distribution between Li and Si projectiles. For Li projectiles, ionized parent and their daughter ions are observed rather strongly in addition to small fragment ions. Intensity distribution of small fragment ions exhibits well-known even–odd oscillations. By contrast, the spectra are completely dominated by small fragment ions for Si projectiles and its intensity distribution follows fairly well the power-law form as described above. Thus, the relative intensity of smallest ions is also expected to reflect well the degree of multifragmentation. It is noted that, for only one-electron loss collisions of Si\(^{4+}\) projectiles, the intensity of the smallest C\(^{+}\) ion occupies about 63\% of the total intensities of small fragment ions C\(^{n+}\) \((n = 1–12)\), implying that a huge amount of energy is certainly deposited into a C\(_{60}\) molecule. Furthermore, a somewhat surprising result is that multifragmentation is also the predominant decay process even for neutral beam incidents of both Li and Si projectiles. This indicates that the interaction region responsible for the ionization (electron loss) of a neutral projectile atom by a neutral molecule is essentially limited to only small impact parameters, where the projectile nuclear charge may be less screened.

3. Calculation method

All the experimental features mentioned above illustrate convincingly that the C\(_{60}\) fragmentation is governed by the amount of excitation energy transferred from the projectiles. It is therefore important to know the deposited energy in individual collision processes in order to understand the ionization and fragmentation mechanisms more systematically.

Theoretical calculations of the total energy deposition into a C\(_{60}\) molecule in a high velocity region have been made by Kabachnik et al. of the Bielefeld group for 0.2–2.0 MeV H\(^{+}\) ions \[10\]. They used the LDA method \[11,12\] with the electron density \(\rho(r)\) in C\(_{60}\) calculated by Puska and Nieminen within the jellium model \[25\]. By assuming straight-line trajectories for projectile ions, the energy deposition for a certain ion trajectory is calculated from

\[
Q(b) = \frac{4\pi Z_1^2}{v^2} \int_{-\infty}^{\infty} dz \rho(r)L(\rho, v),
\]

where the z-axis is the beam direction, \(b\) is the impact parameter measured from the center of the C\(_{60}\) molecule with a relationship of \(|r|^2 = b^2 + z^2\) and \(L(\rho, v)\) is the stopping number. With the aids of the Fermi velocity \(v_F = \left(\frac{3\pi^2\rho}{m}\right)^{1/3}\) and the local plasma frequency \(\omega_p = \left(4\pi\rho\right)^{1/2}\), the stopping number can be approximated by the following analytical expressions \[26\]: 

(i) for \(v \gg v_F\)

\[
L(\rho, v) = \ln\left(\frac{2\pi^2}{\omega_p}\right) - \frac{3}{2}\left(\frac{v_F}{v}\right)^2,
\]

and (ii) for \(v \ll v_F\).
with
\[
C(\chi) = \frac{1}{2(1 - \chi^2/3)^2} \times \left[ \ln \left( \frac{1 + 2\chi^2/3}{\chi^2} \right) - \frac{1 - \chi^2/3}{1 + 2\chi^2/3} \right]
\]
and \( \chi = (\pi v_F)^{-1} \). With these formulae the stopping cross section for \( C_{60} \) is evaluated by
\[
S_{60} = 2\pi \int_0^\infty bQ(b) \, db.
\] (5)

Calculated stopping cross sections, in units of \( 10^{-13} \) eV cm\(^2\)/molecule, are shown by a solid line in Fig. 3 [10]. It is interesting to compare these results with widely used SRIM calculations [27]. To do this, the SRIM stopping cross sections per C atom are simply multiplied by 60 using the Bragg’s additivity rule. The results are given with a dashed line in the figure. Obviously, two results are in reasonably good agreement with each other. The discrepancy between them is found to be only several % over the proton impact energy range investigated. We used this Bragg’s additivity rule in the following calculations of the energy loss in electron loss collisions of Li and Si ions.

Denoting by \( Q_{qk}(b) \) the impact-parameter dependent stopping function for a \( q \rightarrow k \) charge-changing collision, the stopping cross section per C atom, \( S_1(qk) \), may be obtained from the same formula as Eq. (5). Unfortunately, there is no work reported so far for such specific quantity of \( Q_{qk}(b) \), so that we simply put \( Q_{qk}(b) = p_{qk}(b)Q(b) \) using the ionization probability \( p_{qk}(b) \) of projectile by a target particle, and the total stopping function \( Q(b) \). In this way, the stopping cross section \( S_1(qk) \) is obtained by
\[
S_1(qk) = 2\pi \int_0^\infty b p_{qk}(b)Q(b) \, db.
\] (6)

The \( b \)-dependent total stopping function \( Q(b) \) for C atom was calculated from the CasP code developed by Grande et al. for fast heavy projectile ions [20,21]. Note that the values of \( Q(b) \) for Li and Si projectiles are normalized so as to give rise to the SRIM total stopping cross sections of 61.4 (Li) and 176 (Si) in units of \( 10^{-15} \) eV cm\(^2\), respectively. The \( b \)-dependent ionization probabilities \( p_{qk}(b) \) are calculated from the tables of SCA calculations given by Hansteen et al. for K-, L- and M-shell electrons by fast protons [19]. Calculations are made for multiple ionization of Li\(^{q+} \) \((q = 0–2)\) and Si\(^{q+} \) \((q = 0–4)\) ions by protons at velocities of 3.38 \( v_0 \) and 1.69 \( v_0 \), respectively.

An outline of the calculation procedure is described below for the case of Si\(^{q+} \) projectiles. The multiple ionization probability \( p_{qk}(b) \) of Si\(^{q+} \) is calculated within the independent electron model [28],
\[
p_{qk}(b) = \sum_{i=0}^{k-q} \binom{M}{i} \binom{L}{j} p_m^i(1 - p_m)^{M-i} p_l^{j}(1 - p_l)^{L-j},
\]
\[
i + j = k - q,
\]
where \( p_m \) and \( p_l \) are the average single ionization probabilities of M- and L-shells, respectively, obtained from s- and p-subshell ionization probabilities with the number of subshell electrons taken into consideration. \( \binom{N}{r} \) is the binomial

![Fig. 3. Stopping cross section of C\(_{60}\) for fast H\(^+\) ions. Solid line is the LDA calculations [10], and dashed line is the SRIM value [27] multiplied by 60.](image)
coefficient, $M$ and $L$ are the number of electrons in M- and L-shells, respectively; e.g. $M = 2$ and $L = 8$ for Si$^{2+}$. In cases of Li$^{2+}$ ions, K-shell ionization is calculated instead of M-shell. All the subshell binding energies of Li$^{2+}$ and Si$^{2+}$ ions are taken from [29].

It is noted that if the probability $p_{qk}$ is small, corresponding stopping cross sections $S_q(qk)$ also become small according to Eq. (6). For Si$^{2+}$ ions, for instance, calculations of Eq. (7) show $p_{12}(b) > 100p_{13}(b)$ at all $b$’s, implying that the energy transfer in single electron loss collisions is more than hundred times larger than that in four-electron loss collisions. Obviously, such a prediction is unrealistic and contradicts actually what is expected form our experimental mass distribution showing larger energy deposition in four-electron loss collisions [15]. In order to arrive at more realistic results, therefore, all the $q \rightarrow k$ probabilities $p_{qk}(b)$ were normalized so as to satisfy the following equation:

$$2\pi \int_0^\infty b p_{qk}^n(b) \, db = C \text{ (const.)}. \quad (8)$$

Here, the normalized probability $p_{qk}^n(b)$ is given by $p_{qk}(b)C/\sigma(qk)$ using an appropriate normalization factor $C$ and a $q \rightarrow k$ charge-changing cross section calculated from Eq. (8) but substituting the real probability $p_{qk}(b)$. Apart from the magnitude of energy deposition, the electron loss cross section $\sigma(qk)$ is in turn connected directly with experimental yields of fragment ions produced in $q \rightarrow k$ collisions.

It is also noted that the present calculation of $p_{qk}$ is made for protons instead of atomic carbons of the same velocity. Ionization of a charged particle, i.e. projectile ion in our case, by a neutral atom is, however, plausibly thought to occur at only small impact parameters. In such collisions, the nuclear charge of the neutral atom would be less screened and the atom is supposed to act as a charged particle with non-zero effective charge $z_\varepsilon$. The ionization probability by such a particle may reasonably be assumed to follow a simple $z_\varepsilon^2$ scaling law [19]. Thus, the desired values of $p_{qk}$ for neutral carbon atom are supposed to be different from those for protons only in magnitude. However, this difference may be neglected according to our normalization procedure expressed by Eq. (8).

Using the stopping cross sections $S_i(qk)$ determined from Eq. (6), the energy deposition $E_{qk}$ per C$_{60}$ molecule is calculated from the following formula:

$$E_{qk} = Z_{qk}^2 S_1(qk) \frac{60}{\pi a^2}, \quad (9)$$

with the molecular radius $a = 6.6$ a.u. Here, the influence of initial–final charge state difference on the stopping cross sections is taken into consideration by introducing a mean charge $Z_{qk}$ in the $q \rightarrow k$ collision. The mean charge $Z_{qk}$ is simply taken as $(q_e + k_e)/2z_e$ with the initial ($q_e$) and final ($k_e$) effective charges and the mean charge ($z_e$) used in the SRIM code. The effective charges $q_e$ and $k_e$ are calculated from a hydrogenic formula $n_i\sqrt{2I_i}$ for $i = q - 1$ and $k - 1$, respectively, where $n_i$ and $I_i$ are the principal quantum number and the ionization potential (a.u.) of the outermost-shell electrons. The SRIM mean charge $z_e$ is determined in an usual way by $\sqrt{S_1/S_1(H)}$, giving rise to $z_e = 2.48$ and 3.51 for 2 MeV Li and Si projectiles.

4. Results and discussion

Fig. 4 shows some examples of the normalized ionization probability $p_{qk}^n(b)$ for Li and Si ions calculated with $C = 1$ a.u. Step-like structures are accounted for by the relative importance of contributions from different shells of the projectile particles. For instance, ionization of a neutral Li$^0$ projectile is composed of two terms arising from 1s and 2s electrons. Note, however, that the step-like profile was not found for two- and three-electron ionization but only for the single ionization probability $p_{01}$, implying the preferential K-shell ionization in comparison with L-shell. Similar arguments can be addressed for L- and M-shell electrons in Si projectiles, too. Also shown in Fig. 4 are the $b$-dependent stopping functions $Q(b)$, or the energy loss function of C atom for 2 MeV Li and Si projectiles. Step-like structure seen in both projectiles is attributed to the energy loss transferred to K- and L-shell electrons of C atom.
The total energy deposition $E_{qk}$, calculated from Eq. (9), for electron loss collisions of 2 MeV Li and Si projectiles are given in Fig. 5 as a function of the incident charge $q$. Here, we simply put the normalization factor $C$ equal to the geometrical cross section ($C = 12.2$ a.u.) of C atom obtained from the atomic data table given by Desclaux [30]. It appears that the energy deposition $E_{qk}$ spreads widely according to different $q \rightarrow k$ loss collisions and increases with increasing final charge $k$. In particular, the values of $E_{qk}$ reaches about 10 keV for multiple electron loss collisions of Si projectiles resulting in $k = 5$, including also single loss ($4 \rightarrow 5$) collisions. For Si projectiles, it is seen that $E_{qk}$ at a fixed final charge $k$ depends only weakly on the incident charge $q$ and shows a hump structure. As another interesting feature extracted from this figure, the amount of energy loss is nearly equivalent to each other between Li and Si projectiles for the same $q \rightarrow k$ collisions except for their $q$-dependence; e.g. see $k = 2$ and 3.

As mentioned in the previous sections, relative intensity of the smallest fragment ion is supposed to reflect well the total amount of energy deposition. In order to see this relationship, calculated data are plotted in Fig. 6 as a function of the relative intensity $Y_1/Y_c$ between $C^{+}$ and all the small fragment ions $C_n^{+}$ observed experimentally [14,15]. For Si projectiles, a remarkably simple relationship can be seen as shown by a dashed straight line, supporting our basic idea about a possibility of extraction of the inelastic energy deposition form the intensity distribution of small fragment ions. A similar straight-line relationship is also found by plotting $E_{qk}$ as a function of the power $\lambda$ as described previously [15].

As for Li projectiles, however, no such simple relationship is found. This may be partly due to a fact that the intensity of $C^{+}$ is no more a good quantity to reflect the amount of energy deposition because of the strong even–odd oscillations as can
be seen in Fig. 2. On the other hand, the present calculation shows equivalent amount of energy deposition for both Li and Si projectiles for the same $q \rightarrow k$ collisions, as mentioned above (Fig. 5). It leads consequently to anticipation that the intensity distribution of fragment ions should be more or less similar to each other. This prediction, however, is not observed experimentally as already mentioned in Section 2 (see $0 \rightarrow 3$ spectra in Fig. 2). Although the origin of this difference is not clear at the present stage, one possible answer may be given from the consideration of the stopping power partition as discussed in brief below.

According to detailed theoretical work by Campbell et al. [31], the $C_{60}$-multifragmentation is governed only by the internal excitation energy rather than the total energy deposition. Thus, the partition rate into excitation and ionization branch plays an important role in the final distribution of small fragment ions. To our best knowledge, however, only a few investigations have been carried out so far of the stopping power partition [16–18]. Miller and Green [17] calculated the partition rates for a collision system of $H^++H_2O$ in the energy range from 0.1 keV to 10 MeV, showing clearly that the partition rate changes significantly depending on the impact velocity. For high velocities ($v > 2.5v_0$), about 10% of the total energy loss is spent for excitation and 90% for ionization. At $v = 1.69v_0$, being equivalent to present Si projectiles, the excitation branch occupies about 20%. If these values are employed straightforwardly in the present work, the internal excitation energy given by Si impact becomes about twice as large as that by Li impact, leading qualitatively to a possible explanation of the present question. To achieve further discussion, more accurate information about the partition rates concerning ion–$C_{60}$ collisions is needed.

We also note briefly about the effect of the nuclear energy loss. In the present collision systems, the stopping power ratios between nuclear and electronic is estimated, from the SRIM code, to be 0.14% and 2.8% for Li and Si projectiles, respectively. Namely, the nuclear stopping for Si is 20 times larger than for the Li projectiles. Such a large difference of nuclear energy loss may also affect the final mass distribution. This speculation may be supported qualitatively by low velocity collision experiments [32] in which mass distribution of fragment ions are significantly different for He, Ne and Ar projectiles.

5. Conclusions

We have investigated the $C_{60}$-multifragmentation following electron loss collisions of 2 MeV Li and Si projectiles. Using a simple calculation method, the electronic energy deposition per $C_{60}$ has been obtained. In the present method the impact-parameter dependent energy loss function $Q_{qk}(b)$ for $q \rightarrow k$ electron loss collisions is replaced by $p_{qk}(b)Q(b)$, where $p_{qk}(b)$ is the projectile ionization probability by $H^+$ obtained from the tables of SCA calculations [19], and $Q(b)$ the total energy loss function for Li (or Si) + C calculated from the CasP code [20,21]. The total energy deposition per $C_{60}$ is calculated using the Bragg’s additivity rule supported by LDA calculation for $H^++C_{60}$ collision [10].

It is found that, in semilogarithmic plot, the calculated energy deposition reveals a linear relationship with the degree of $C_{60}$-multifragmentation
for Si projectiles, while this is not the case for the Li incidence. We also found that the energy deposition is nearly equivalent for both projectiles for the same $q \rightarrow k$ loss collisions, which is, however, inconsistent with previous experimental results indicating that a larger amount of internal energy is certainly deposited by Si impacts. This discrepancy may be attributed to the different partition rates for these projectiles. In order to understand the energy loss mechanism more precisely, it is urgently important to study the stopping power partition in ion–C$_{60}$ collisions.

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References