Disintegration of swift carbon clusters during passage through matter

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Thin luminescent foils covered upstream with layers of Formvar or gold of various thicknesses were bombarded with C_8 clusters with energies between 5 and 10 MeV. The C_8 induced relative luminescence yield Φ_8 increases with growing layer thickness and approaches smoothly the value $8\Phi_1$, i.e., the luminescence yield induced by eight well-separated C_1 cluster constituents. This is a clear demonstration of the gradual separation between the cluster constituents during the passage of the cluster through the layers. The layer thickness necessary to separate cluster constituents far enough to act as independent atomic ions with respect to the luminescence process was calculated. The result is that Formvar layers with a thickness of about 1150 nm or $\approx$250-nm-thick gold layers are necessary to establish this mutual independence of the cluster constituents. Both calculated values agree roughly with the layer thickness obtained from an extrapolation of the experimental data.

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I. INTRODUCTION

Polyatomic projectiles such as carbon clusters C_n with $n>1$ deposit a huge amount of energy within a small volume along the track when they penetrate matter. The energy density along the track is comparable with, or even larger than energy densities accessible with, the fastest and heaviest atomic projectiles from the big heavy ion facilities. This can be concluded, e.g., from the observation that the dimensions of craters generated by 20 MeV C_{60} cluster ions hitting solid matter exceed those produced by GeV atomic gold ions [1] or from the observation that 18 MeV C_{60} clusters create continuous tracks in metals with considerably larger diameters than atomic projectiles such as Pb or U [2]. Likewise, the fact that MeV clusters are able to sputter large size molecules with yields exceeding those obtainable with MeV atomic ions has to be attributed to the much larger energy density created along the cluster trajectory [3]. Another example for the high energy density is the huge defect production in crystalline high-T_c superconductors observed after the impact of C_{60} ions. Significantly more defects are produced if the superconductor is bombarded with MeV cluster ions instead of GeV atomic ions [4].

Swift cluster ions are therefore suited in all cases where the deposition of very high energy densities is required. As an application, the use of cluster ions for the indirect drive of inertial fusion was proposed [5]. During the passage through matter, however, the distance between the cluster constituents increases continuously due to their interaction with the electrons and nuclei of the surrounding atoms and due to their mutual Coulomb repulsion or due to the relaxation of excited states. As a result, the deposited energy density decreases. Therefore, the advantage of swift cluster ions as a source of high energy density persists only within the small distance traveled by the cluster constituents in close proximity. The aim of this work was to determine these distances in matter, and to demonstrate the gradual separation between the constituents during the passage of the cluster through different materials. We chose Formvar and gold as examples for materials with low and high nuclear charge *Z*.

II. EXPERIMENT

For the present investigation, we make use of the fact that the relative luminescence yield Φ_n measured with carbon clusters C_n hitting a thin luminescent sample, is significantly smaller than the *n*-fold yield Φ_1 induced by the monoatomic cluster constituent C_1 at the same velocity as C_n. The data obtained from luminescence experiments with a 100-nm-thin POPOP sample [6] show that the ratio $\Phi_n/n\Phi_1$ is always smaller than 1 for $n>1$. A value of 1 is therefore expected, if the n cluster constituents hit the luminescent sample well separated, i.e., if the distances between the constituents are large enough for all of them to act as independent C_1 projectiles with respect to the luminescence process. The necessary separation can be achieved in an additional layer of appropriate thickness which covers upstream the luminescent sample. The increase of the luminescence yield Φ_n with growing layer thickness is then a measure for the disintegration of the cluster C_n in the additional layer. The latter, of course, has to be nonluminescent.

Self-supporting $\approx$200-nm thick foils consisting of a 3:1 (w/w) mixture of Formvar and POPOP were used as luminescent samples and covered with Formvar or gold layers of various thicknesses (up to 620 nm and 190 nm, respectively).

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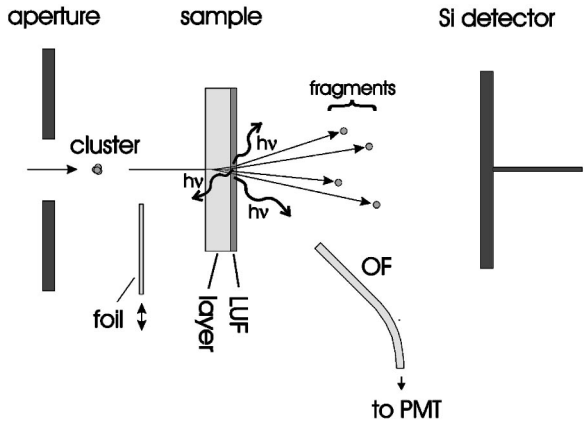

FIG. 1. Experimental scheme; LUF: luminescent foil; $h\nu$: photons produced in the LUF; OF: optical fiber; PMT: photomultiplier outside the recipient.

The samples were moved into the ion beam at exactly the same position and irradiated by C_8 clusters from the Erlangen tandem accelerator with energies between 5 and 10 MeV and beam intensities of a few hundred particles per second. In these samples, the induced luminescence originates from the fluorescence of the POPOP molecules, the spectrum of which extends from about 400 to 550 nm [7]. Luminescence photons were collected by two optical fibers and transferred to a cooled photomultiplier outside the vacuum chamber as indicated in the scheme of Fig. 1. Cluster fragments traversing the foil were detected by a Si-surface barrier detector. The photons were recorded by time-correlated single-photon counting using the response from both detectors [6].

The thickness of the luminescent foils and their additional Formvar and gold layers was determined from the energy loss of α particles or C_1 ions and from light absorption measurements. It was checked that neither Formvar nor gold layers emit photons upon cluster impact in the accessible spectral range (300–750 nm). An additional thin Formvar foil could be moved into the beam in front of the samples (about 5 mm upstream and about 100 nm thickness). With this foil in the beam, C_8 clusters break apart into their eight constituents that then hit the sample well separated in space. The photon yield Φ_8 observed in this case was $8\Phi_1$.

III. EXPERIMENTAL RESULTS

The luminescence yields Φ_8 and Φ_1 obtained with C_8 clusters and monoatomic C_1 projectiles, respectively, were measured as a function of the thickness of the additional layers on top of the luminescent foil. The results are presented as normalized yields $\Phi_8/8\Phi_1$ in Figs. 2 and 3. The ratio $\Phi_8/8\Phi_1$ increases clearly with growing thickness d_F or d_{Au}. This increase is unambiguously due to the gradual separation between cluster constituents during the passage of the cluster through the additional layers. After about 600 nm of Formvar or 200 nm of gold, the separation has proceeded so far that Φ_8 reaches $\approx$80% of the yield obtained when eight well-separated carbon ions hit the bare luminescent foil. The observation that a much thinner gold layer causes a separation between the constituents similar to that caused by

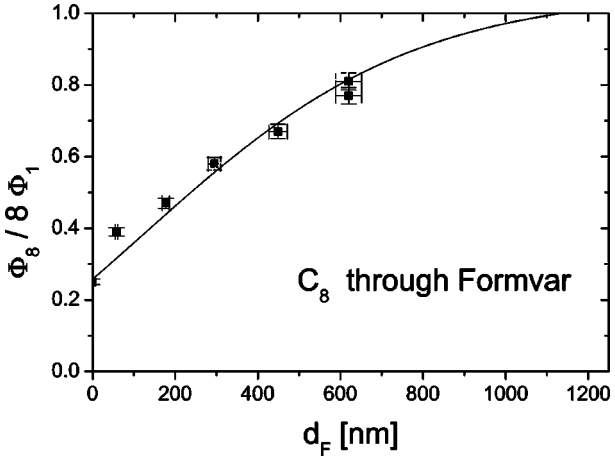

FIG. 2. Normalized luminescence yield $\Phi_8/8\Phi_1$ as a function of the Formvar layer thickness d_F on top of a 200-nm-thick luminescent Formvar(POPOP) foil. Φ_8 and Φ_1 are the relative luminescence yields induced by C_8 and C_1 ions, respectively, both with the same velocity. The cluster energy was varied between 5 and 10 MeV (see text). The solid line represents an extrapolation to larger values of d_F.

a much thicker Formvar layer is well explained by the higher-Z value of gold which leads to an enlarged contribution of nuclear processes. The Formvar result is consistent with the observation that the yield of H^+ ions, desorbed from the backside of carbon foils under the bombardment with 10 MeV C_{10} clusters, is about ten times larger than the yield obtained with 1 MeV C_1 ions, when the foil thickness reaches a value of about 900 nm [8].

It should be noted that the initial energies of the C_8 clusters were chosen in such a way that the energy of the cluster constituents exiting the additional Formvar layer was roughly 700 keV independent of the layer thickness. Each time a different sample was used, the terminal voltage of the accelerator was varied until the energy signal of the Si-detector was identical with the signal observed when the bare luminescent foil was bombarded. This precaution was taken

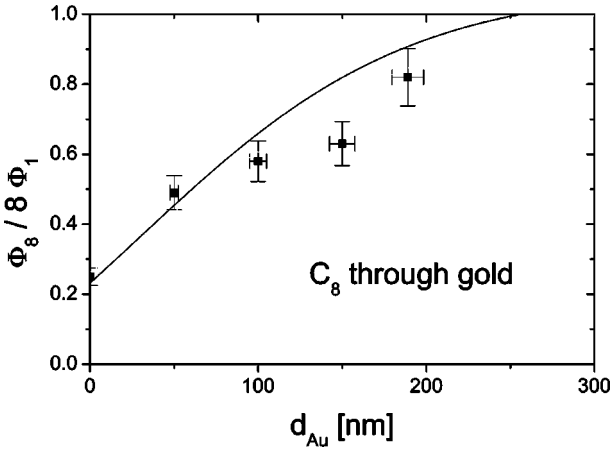

FIG. 3. $\Phi_8/8\Phi_1$ as a function of the gold layer thickness d_{Au} on top of the luminescent foil. The cluster energy was 7.8 MeV. The solid line represents an extrapolation to larger values of d_{Au}.

in order to eliminate a possible energy dependence of the luminescence yield. But an investigation of this dependence performed later with C_8 clusters in the energy range 5–10 MeV and with C_1 ions between 0.625 and 1.25 MeV using a bare luminescent foil showed that the yield remains constant in this energy region. Therefore, the measurements with gold samples were carried out with the constant cluster energy of 7.8 MeV. The energies of the cluster constituents exiting the gold layers varied between 700 and 1,000 keV in this case.

IV. AVERAGE SEPARATION BETWEEN CLUSTER CONSTITUENTS

A. Calculation with the TRIM code

TRIM calculations with the code SHRIM-2000.40 [9] were performed to get a rough idea for the average separation distance $\langle \delta \rangle$ between cluster constituents exiting a 600-nm Formvar layer with an energy of 700 keV. The code allows us to calculate the trajectory of an atomic ion traversing a thin sample and yields the coordinates of the ion at the exit side. Only the interactions between the atomic ion and target atoms are considered in these calculations. They do not include typical cluster features such as (i) mutual Coulomb repulsion between cluster constituents, (ii) influence of neighboring constituents on the charge state of a single constituent, and (iii) the sweeping-out-electrons effect, which means that a constituent travelling in the wake of the constituents ahead of it may interact with a smaller electron density [10]. Thus, the exit coordinates of C_1 ions hitting the sample one after the other are actually calculated and not the coordinates of cluster constituents simultaneously impinging onto the sample.

In order to find out, if the lack of typical cluster features yields unrealistic average separation distances $\langle \delta \rangle$, the result of a TRIM calculation was compared with the prediction of a more realistic model [11] developed recently to describe the passage of clusters through matter. This model includes, e.g., the mutual Coulomb repulsion of the constituents. The average distance $\langle \delta \rangle$ predicted by this model between cluster constituents at the exit of a 40 $\mu g\,cm^{-2}$ carbon foil is about 1 nm for C_n clusters with $n<10$ and an initial energy of 2 MeV per carbon atom [11]. The average distance $\langle \delta \rangle$ calculated by TRIM with the same parameters as described above is 1.3 nm. The agreement between both results is quite satisfactory and seems to indicate that the cluster features not considered in TRIM are only of minor importance. It shows further that the TRIM results are obviously not unrealistic.

The TRIM calculations were performed for a large number of C_1 ions exiting a 600-nm-thick Formvar layer with an energy of 700 keV. The calculations were actually performed for two groups of C_1 ions hitting the layer at a distance of 0.13 nm in order to simulate the impact of two adjacent constituents of a C_8 cluster with an assumed linear shape. Recent investigations of cold C_8 clusters showed that two of three clusters have a linear shape [12] with a bonding length of about 0.13 nm [13]. This choice of two groups of C_1 ions, starting 0.13 nm apart, neglects that C_8 clusters exhibit also an annular shape and that they hit the Formvar surface with a statistically distributed orientation. Besides this, clusters under the conditions of the presented experiments are most probably not cold. The TRIM calculations showed, however, that the absolute values of the coordinates at the exit side are more than one order of magnitude larger than the cluster bonding length, which means that both the structure of the cluster and its orientation relative to the front surface do obviously not have a large influence on the calculated results.

After the passage of 600 nm Formvar, the average distance $\langle \delta_F \rangle$ between exit points of ions belonging to the two groups of constituents turned out to be ≈ 21 nm. An inspection of the C_1 trajectories shows that the ions spread out on their way through the Formvar layer gradually in the transverse direction due to successive small angle scattering. Occasional large angle scattering events are rather scarce (five out of 1000 ions are scattered through more than 20°). A similar result can be expected for C_8 clusters passing through Formvar. The transverse spread might, however, be slightly more pronounced due to the mutual Coulomb repulsion of the cluster constituents. A calculation for C_1 ions exiting a 200-nm-thick gold layer with an energy of 700 keV yields an average separation distance $\langle \delta_{Au} \rangle$ of 32 nm. The fact that similar values are obtained for $\langle \delta_F \rangle$ and $\langle \delta_{Au} \rangle$ is not surprising, because the ratio $\Phi_8/8\Phi_1$ is approximately the same in both the cases.

B. Calculation via the interaction range

After the passage through 600-nm Formvar or 200-nm gold, the cluster constituents are still not separated far enough from each other to act as independent C_1 ions with respect to the luminescence process. The $\Phi_8/8\Phi_1$ ratios measured at these thicknesses are smaller than 1, i.e., C_1 ions then entering the luminescent foil can still interact with each other. It would be interesting to know the approximate Formvar or gold thickness at which a complete separation with respect to the luminescence process is achieved. An extrapolation of the experimental data presented in the Figs. 2 and 3 yields only a rather rough value for this thickness, because the precise shape of the $\Phi_8/8\Phi_1$ versus d_F (or d_{Au}) function is not known. Therefore, it seemed to be more reliable to estimate the smallest separation distance $\langle \delta_{min} \rangle$ instead which assures that 700 keV C_1 ions entering the luminescent foil behave as independent particles. Subsequently, the layer thickness that causes an average separation equal to $\langle \delta_{min} \rangle$ can be deduced from TRIM calculations.

The necessary criterion for mutual independence with respect to the luminescence process is that the energized volumes along the track of each single cluster constituent do not interact with each other. This means that the average distance between cluster constituents at the entrance of the luminescent foil has to be larger than $2R_m+D$. The quantity R_m is the radius of the volume energized by secondary electrons along the C_1 track (assumed to have a cylindrical shape) and D is the interaction range between two excited POPOP molecules, i.e., the smallest distance for which quenching of the luminescence, on an average, does not exist. In the following, we present estimations of both quantities.

1. Radial extension of the energized volume

For the cluster energies in question, the energy deposition in the sample occurs mainly via electronic interactions. The

extension of the energized volume perpendicular to the track is determined by the range R of the electrons that are set free in ionization processes, if only electrons are assumed to transport the energy lost by the projectile into the surrounding solid. The range of these electrons in a solid depends on the energy E_e transferred to the electrons in a binary collision with a cluster constituent of energy E_0:

$$E_e = \frac{4m_e}{M} E_0 \cos^2\varphi, \tag{4.1}$$

where φ is the scattering angle of the recoiling electron in the laboratory system and m_e and M the mass of the electron and the C_1 ion, respectively. The maximum energy transferred to electrons by 700 keV C_1 ions (typical energy of the C_1 ions entering the luminescent foil) is $E_e = 128$ eV. Unfortunately, there exist no range data for electrons entering a Formvar(POPOP) foil with such small energies.

Therefore, electron ranges in polystyrene were used. They were obtained from energy loss data for low energy electrons ($10-10^4$ eV) in polystyrene calculated by Ashley *et al.* in the model insulator theory [14]. The electron range R is related to the energy loss dE/dx by

$$R = \int_{2.7\ \mathrm{eV}}^{E_e} \left(\frac{dE}{dx}\right)^{-1} dE. \tag{4.2}$$

The lower limit of 2.7 eV was chosen, because the mean excitation energy of a POPOP molecule is 2.7 eV, i.e., electrons with this energy can still excite POPOP molecules. In solving the integral, it was assumed that the integrand in Eq. (4.2) is zero for $E_e = 0$ and that a linear interpolation to its value at 10 eV can be made [15]. With the crude assumption that the electrons travel along a straight line in the direction of the scattering angle φ, the positions of all electrons with the final energy of 2.7 eV were calculated for all scattering angles between 0° and 90° degrees. The maximum distance of all these positions from the ion trajectory, i.e., the radial extension R_m of the energized volume, turns out to be 14 nm. Accordingly, electrons that are set free in collisions with 700 keV C_1 ions are found in a cylindrical volume with a diameter of about 28 nm centered around the ion track.

2. Interaction range between excited POPOP molecules

Excited POPOP molecules interact with each other mainly via dipole-dipole interaction. This interaction leads to exciton-exciton annihilation processes resulting in a reduced luminescence yield in favor of radiationless electronic transitions [16,17]. It is also possible that further electrons are set free in these processes [18]. The properties of the electronic states involved, i.e., mobility and lifetime of the excitons, and the density of the luminescence centers are the decisive factors that determine the range of this interaction.

A rough idea for the interaction range D between excited POPOP molecules can be derived from luminescence data obtained recently with high-energy atomic ions [19]. These data show that the energy density ϵ deposited along the ion track is the relevant parameter for the luminescence yield and that the luminescence originates only in regions with $\epsilon \leqslant 0.017$ eV nm^{-3}. This means that the immediate vicinity of the ion track does generally not contribute to the luminescence due to its high-energy density. The largest ϵ value ($\epsilon = 0.017$ eV nm^{-3}), for which quenching does not exist according to the luminescence data, for example requires the presence of 6.3×10^{18} excited POPOP molecules within 1 cm^3, because the mean excitation energy of the molecule is 2.7 eV. The distance between these excited molecules is 5.4 nm, if they are assumed to be equally distributed in space. This result can be regarded as a reasonable estimation for the interaction range D between two excited POPOP molecules, i.e., for the minimum distance at which luminescence quenching, on an average, does not occur. The consequence for the present cluster experiment is that a mean distance of more than 5.4 nm should exist between the energized zones of two adjacent cluster constituents.

3. Layer thickness for complete separation

According to the discussion presented above, the mean separation distance between the trajectories of adjacent constituents entering the luminescent foil has to be at least 33.4 nm to assure mutual independence of the C_1 ions. The Formvar layer thickness necessary to achieve this separation $\langle \delta_{min} \rangle$ of 33.4 nm has been deduced from TRIM calculations. For this purpose, the exit coordinates of C_1 ions traversing layers of various thicknesses with an exit energy of 700 keV were calculated along the lines given above. It turned out that a thickness between 1100 and 1200-nm is necessary for layers of Formvar. If the C_8 cluster ions pass through such layers, the luminescence ratio $\Phi_8/8\Phi_1$ should be 1. Inspection of Fig. 2 shows that a smooth curve is obtained, if the data points are extrapolated to $\Phi_8/8\Phi_1 = 1$ at $d_F \approx 1{,}150$ nm. Thus, it seems reasonable to state that a Formvar thickness of roughly 1150 nm is necessary to separate C_1 ions with an exit energy of 700 keV far enough from each other so that they act as independent ions with respect to the luminescence process. A TRIM calculation performed, for the same purpose, for gold layers showed that ≈ 250 nm thickness of gold are necessary to obtain a separation of 33.4 nm. Again, the extrapolation of the experimental data yields a reasonable smooth curve, if the ratio $\Phi_8/8\Phi_1 = 1$ at $d_{Au} = 250$ nm is included (see Fig. 3).

V. CONCLUSION

In conclusion, it was found by means of luminescence experiments that the distance between cluster constituents increases gradually, when clusters pass through matter. From these experiments, it could be deduced that C_8 clusters with energies between 5 and 10 MeV have to traverse roughly 1150 nm of Formvar or 250 nm of gold, before the cluster constituents are separated far enough from each other in order to act as independent carbon ions with respect to the luminescence process in a thin Formvar(POPOP) foil. The average separation distance between adjacent constituents at the exit side of these layers is ≈ 33 nm as obtained from TRIM calculations. An estimate of both the radial extension of the energized volume and the interaction range between two

excited POPOP molecules yields a distance between two adjacent constituents of at least 33 nm in order to assure that the energized volumes along the ion tracks, on an average, do not interact with each other. It follows from this investigation that the very high-energy density along the cluster trajectory, which is a typical feature of the cluster-solid interaction, persists only within the first 1150 nm of Formvar or 250 nm of gold in the present case.

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