Coulomb Explosion Patterns of Fast C_{60} Clusters in Solids

You-Nian Wang* and Hua-Tan Qiu

The State Key Laboratory of Materials Modification by Laser, Electron, and Ion Beams, Dalian University of Technology, Dalian 116023, People's Republic of China

Z. L. Mišković

Department of Applied Mathematics, University of Waterloo, Waterloo, Ontario, Canada N2L 3G1

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The molecular dynamics method is used to simulate Coulomb explosion of fast C_{60} ion clusters in an Al target, taking into account dynamical screening of interionic interactions by the electron gas of the target. It is found that the wake forces in the medium are strong enough, depending on cluster speed, to stabilize the whole cluster against Coulomb explosion, or to compress the trailing part of the cluster, for prolonged times of penetration through the target. This is encouraging news for such cluster-ion beams applications where massive energy depositions in small volumes of targets are desired.

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Coulomb explosion of large atomic clusters has become the focus of many recent investigations in diverse areas. For example, high-energy ions are produced in explosions of very large clusters, heated with high-intensity femtosecond laser pulses, which may be used to create deuterium plasmas with sufficient ion temperatures for substantial nuclear fusion in a tabletop experiment [1]. Furthermore, with recent advances in accelerator techniques for production of fast beams of large clusters [2], at the speeds in excess of Bohr velocity v_B, a renewed interest arises in the areas of cluster interactions with the solid and hot plasma targets. It is expected that a massive energy deposition introduced by large clusters in a small volume of the target may give rise to qualitatively new effects in such applications as solid state detectors [2], materials modification [3], or inertial confinement fusion in large-scale facilities [4]. Interaction of fast cluster-ion beams with such targets is dominated by the response of target electrons, which turns out to be quite different from the response to ordinary ion beams.

Upon entering a solid target, an atomic cluster with the speed $v > v_B$ will lose its binding valence electrons after traversing a few atomic layers, and thus emerging ions will attain equilibrium charge states within about 1 fs, while still retaining the initial geometry of the cluster. Further penetration of the ion cluster is accompanied by electronic excitations of the target, which exhibit strong interference due to spatial correlation among the ions. As long as the interionic distances within the cluster are smaller than the characteristic length scale of the electronic excitation pattern, a constructive interference gives rise to enhanced energy losses to the target electrons, when compared to the energy losses of individual ions moving at the same speed [5]. While this energy-loss enhancement is certainly an important feature for the above mentioned applications of cluster-ion beams, one expects that an increase of interionic distances in the course of penetration, due to Coulomb repulsion among the ions, may soon switch the interference effect to a destructive regime accompanied by a reduction of cluster energy losses [6]. It is therefore important to understand the peculiarities of Coulomb explosions of large clusters in solids and their role in energy deposition in the target.

Coulomb interactions among constituent ions of a fast cluster are screened by the collective response of the electron gas in the target in a manner which depends on the cluster speed v [7]. A common qualitative measure of such a dynamic screening of the Coulomb interaction between two ions with charges q_1 and q_2, moving at equal velocities $\mathbf{v}$ at a distance r_{12} from each other, is provided by the empirical Yukawa-type potential $(q_1 q_2/r_{12})\exp(-r_{12}/\ell)$ with the screening length $\ell = v/\omega_p$, where ω_p is the plasma frequency of the electron gas. This model potential has been frequently used in most of the theoretical accounts of the Coulomb explosion of *small* clusters [8–10] and in a few existing numerical approaches to the Coulomb explosion of *large* clusters in solids [11,12]. The Yukawa-type interaction potential clearly shows that the Coulomb explosion of a cluster is less vigorous in a solid target than in a free space, with the ion debris entering the ballistic regime of expansion as soon as the interionic distances exceed $\ell = v/\omega_p$. However, a more realistic interaction between the ions moving in a solid is far from being radially isotropic, as given by the Yukawa potential. It has long been known [13] that the collective response of the target electrons to a *single* fast ion gives rise to an oscillatory electron density polarization, with the characteristic wavelength $\lambda = 2\pi v/\omega_p$, which has the form of a cylindrically symmetric stationary wake trailing behind the ion. Coulomb explosion studies of fast *diatomic* molecules in solid targets have shown that the dynamically screened interaction between the two ions is strongly influenced by the interference of their wakes, exhibiting a pronounced tendency to align the molecular axis in the direction of

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motion [14,15]. One may then expect that the dynamically screened Coulomb forces, acting at different points in a *large* cluster, will also be strongly asymmetric relative to the direction of cluster motion. In particular, the forces on the leading and trailing ions, or on the ions in the lateral positions, may be quite different, leading to anisotropic Coulomb explosion patterns, which are explored in this Letter for the first time.

We choose C_{60} as a model cluster because, in a Coulomb explosion, its equilibrium spherical-shell structure will be highly susceptible to any asymmetry in the interionic interactions. Moreover, significant progress has been achieved in recent experimental studies of the interaction of energetic C_{60} clusters with solids. The Orsay group was the first to measure the energy losses of the energetic C_{60} clusters in the energy range from 6 to 30 MeV in thin carbon foils [2]. Also, the track formation has been observed experimentally in metals, insulators, and semiconductors under the irradiation by fast C_{60} cluster beams [3]. On the theoretical side, in a recent molecular-dynamics (MD) simulation of the interaction of a fast C_{60} cluster with an electron gas, the radially isotropic Yukawa potential was used to drive the Coulomb explosion, giving rise to expanding spherical-shell patterns [12]. On the other hand, wake effects have been taken fully into account in recent self-energy calculations for a randomly oriented C_{60} ion cluster in an Al target, showing that the initial potential energy, stored in the cluster upon its ionization, takes negative values for cluster speeds up to about $5v_B$ [16]. This unexpected result was taken as an indication that the whole cluster, or parts of it, may remain stable against the Coulomb explosion in a solid target [16].

We use the MD method to simulate Coulomb explosion patterns of a fast C_{60} cluster entering an Al target with the initial speed v_0 of the order of several v_B. The dynamically screened Coulomb interactions between the ions within the cluster are derived by means of the linear-response dielectric theory [13], which is expected to be adequate for such high speeds [4,5]. Generally, the stopping forces $\mathbf{F}_s$ due to electron excitations in the target, the dynamically screened Coulomb interaction forces $\mathbf{F}_{\rm int}$, and the random-deflection forces due to elastic multiple scattering at the target atoms all act simultaneously upon the individual carbon ions in the course of penetration. Compared with the other two forces, the multiple-scattering effect is not significant in the high-speed regime and can therefore be neglected in the present approach [8,17]. The equation of motion for the ith ion within the cluster is then given by

$$m\, d\mathbf{v}_i/dt = \mathbf{F}_s(\mathbf{v}_i) + \sum_{j(\neq i)=1}^{60} \mathbf{F}_{\rm int}(\mathbf{r}_{ij}, \mathbf{v}_i, \mathbf{v}_j)\,, \quad (1)$$

for $i = 1, 2, \ldots, 60$, where $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$ is relative position vector between the ith and the jth ion, and m is the carbon atomic mass. The stopping forces and the interaction forces are obtained from the interionic interaction potential $U(\mathbf{r}_{ij}, \mathbf{v}_i, \mathbf{v}_j)$, respectively, as

$$\mathbf{F}_s(\mathbf{v}_i) = -\partial U(\mathbf{r}_{ij}, \mathbf{v}_i, \mathbf{v}_i)/\partial \mathbf{r}_i|_{\mathbf{r}_{ij}=0}\,, \quad (2)$$

$$\mathbf{F}_{\rm int}(\mathbf{r}_{ij}, \mathbf{v}_i, \mathbf{v}_j) = -\partial U(\mathbf{r}_{ij}, \mathbf{v}_i, \mathbf{v}_j)/\partial \mathbf{r}_i\,, \quad (3)$$

with

$$U(\mathbf{r}_{ij}, \mathbf{v}_i, \mathbf{v}_j) = \int d\mathbf{r}\, \rho_{\rm ext}(\mathbf{r} - \mathbf{r}_i - \mathbf{v}_i t) \times \Phi(\mathbf{r} - \mathbf{r}_j - \mathbf{v}_j t)\,, \quad (4)$$

where $\rho_{\rm ext}(\mathbf{r} - \mathbf{r}_i - \mathbf{v}_i t) = Ze\delta(\mathbf{r} - \mathbf{r}_i - \mathbf{v}_i t) - e\rho_e(\mathbf{r} - \mathbf{r}_i - \mathbf{v}_i t)$ is the charge distribution of the ith carbon ion, with $Z = 6$ being the atomic number of carbon and $\rho_e(\mathbf{r} - \mathbf{r}_i - \mathbf{v}_i t)$ being the distribution of the electrons still bound to the ith ion. Here, $\Phi(\mathbf{r} - \mathbf{r}_j - \mathbf{v}_j t)$ is the scalar potential of the jth ion moving in the electron gas, with the space-time Fourier transform readily obtained from the Poisson equation, as follows:

$$\Phi(\mathbf{k}, \omega) = \frac{8\pi^2 e}{k^2} \frac{Z - \rho_e(\mathbf{k})}{\varepsilon(k, \omega)} e^{-i\mathbf{k}\cdot\mathbf{r}_j} \delta(\omega - \mathbf{k}\cdot\mathbf{v}_j)\,, \quad (5)$$

where $\rho_e(\mathbf{k})$ is the space Fourier transform of the electron distribution $\rho_e(\mathbf{r})$ and $\varepsilon(k, \omega)$ is the longitudinal dielectric function of the electron gas.

We take all the charge states of carbon ions in the cluster to be equal and constant during the Coulomb explosion, with the value determined by the equilibrium charge state of a single carbon ion moving through the Al target at the same speed as the cluster ions. This value is obtained, in a consistent manner, by using a variational treatment of the bound electron distribution $\rho_e(r)$ with an exponential screening, subject to a velocity-dependent electron stripping criterion [18]. For example, the ionizations of carbon ions, thus obtained, take the values $q = 0.4085$, 0.5427, and 0.6459 for the ion speeds $v = 2$, 3, and 4 (in v_B), respectively. It should be mentioned, however, that the ionization degree of each ion in the cluster may be reduced to some extent by the vicinity of the other ions via the interaction potentials $U(\mathbf{r}_{ij}, \mathbf{v}_i, \mathbf{v}_j)$ [19]. A proper treatment of this effect in a large cluster would require a modification of the MD method to solve the equations of motion for ions, coupled with a self-consistent procedure of obtaining their charges [20], which lies beyond our current computational capacity.

A dielectric description of wakes in solids may be formulated by means of a dielectric function which includes a response of both the valence and the core electrons of the target [21]. For a moderately high speed v of cluster ions, the interaction range for a core-electron excitation v/ω_c (ω_c being the corresponding excitation frequency [22]) is much shorter than the interionic distances within the cluster, while the interaction range for the collective response of quasifree gas of valence electrons, v/ω_p, is comparable to those distances. It is clear that the interionic

interactions $\mathbf{F}_{\text{int}}$ are dominated by the response of valence electrons, while the core-electron excitations may only modify, to some extent, the stopping $\mathbf{F}_s$ of individual ions in the cluster, as shown in Ref. [22]. Since modifications of stopping forces influence only the motion of the cluster center of mass, our modeling of Coulomb explosion patterns employs the so-called plasmon-pole approximation [13] for the dielectric function $\varepsilon(k, \omega)$ in (5), which takes full account of the long-ranged wake pattern of the response of valence electrons. It follows then that the interionic forces, obtained from Eqs. (3)–(5), in general violate Newton's third law, that is, $\mathbf{F}_{\text{int}}(\mathbf{r}_{ij}, \mathbf{v}_i, \mathbf{v}_j) \neq -\mathbf{F}_{\text{int}}(\mathbf{r}_{ji}, \mathbf{v}_j, \mathbf{v}_i)$, as a direct consequence of the wake effects in the medium. To illustrate this, we consider two ions moving through an Al target at equal speeds of $4v_B$ with the relative position $\mathbf{r}_{ij} = (X, Y, Z)$ given in a coordinate frame with the Z axis along the direction of motion. Figure 1 shows asymmetric dependences on Z, with $X = Y = 0.3$ nm, of the longitudinal and transversal components, F_z and F_y, of the force $\mathbf{F}_{\text{int}}$.

Having defined all of the ingredients of the theory, we solve the equations of motion (1) for all ions in the cluster ($i = 1, 2, \ldots, 60$), with the set of initial positions $\mathbf{r}_i(0) = \mathbf{r}_{i0}$, where $\mathbf{r}_{i0}$ are the equilibrium positions of carbon atoms in C_{60}, and with all the initial velocities being equal to the initial cluster velocity, $\mathbf{v}_i(0) = \mathbf{v}_0$. It should be noted that the velocities of individual ions deviate only slightly from the initial velocity, $|\mathbf{v}_i - \mathbf{v}_0| \ll v_0$; that is, the Coulomb explosion proceeds in an adiabatic manner in a moving frame of reference attached to the cluster center of mass. Figure 2 shows several snapshots of the ion positions in this frame, at times $t = 5$, 10, 15, and 25 fs, for a C_{60} cluster entering an Al target with the speed $v_0 = 4v_B$. The patterns appear to be cylindrically symmetric about the direction of motion and are changing on a time scale slower than fs. The initial spherical shell of C_{60} expands into a mushroom-, or rather, a jellyfishlike shape after 25 fs, with a somewhat unexpected rearrangement of ion positions. While the leading group of about two-thirds of all ions is expanded to about twice the equilibrium size of C_{60}, the trailing one-third of ions remain bunched in a small volume. This compression of the trailing part of the cluster is clearly a consequence of interfering ion wakes of the leading part. For the sake of comparison with the results in Fig. 2, let us mention that a modeling of Coulomb explosion driven by Yukawa-type interionic interactions [12,16] would give a sphere of about 5.6 nm in diameter after 25 fs, which is about 8 times the equilibrium diameter of C_{60}. Thus, a proper treatment of dynamically

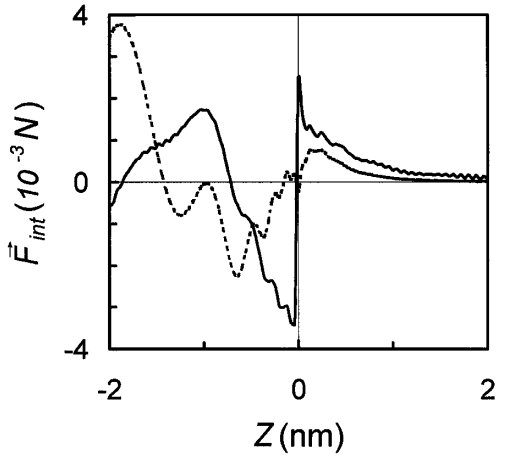

FIG. 1. Components of the interaction force $\mathbf{F}_{\text{int}}$ between two ions moving through an Al target with equal speeds of $4v_B$ at a relative position (X, Y, Z) from each other, with the Z axis in the direction of motion. The dependences on Z (in nm) of the longitudinal F_z (solid line) and transversal F_y (dashed line) force components (in 10^{-2} N) are shown for $X = Y = 0.3$ nm.

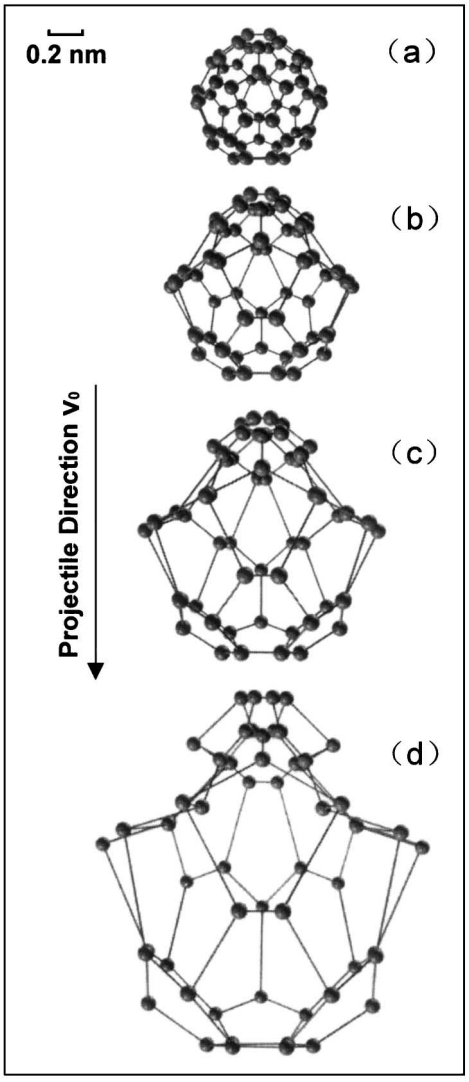

FIG. 2. 3D Coulomb explosion patterns of C_{60} moving through an Al target at speed $v_0 = 4v_B$ in the indicated direction. Snapshots of ion positions are given in a frame of reference attached to the cluster, for several penetration times: (a) $t = 5$ fs, (b) $t = 10$ fs, (c) $t = 15$ fs, (d) $t = 25$ fs.

screened Coulomb interactions exhibits a dramatic slowing down of Coulomb explosions of large clusters in solid targets, as compared to empirical potentials. Moreover, other runs of our MD simulation show that the wake effects on Coulomb explosion patterns strongly depend on cluster speed. For example, at a lower speed of $v_0 = 2v_B$, the whole C_{60} cluster remains quite stable against Coulomb explosion, with the bucki-ball shape turned into a pearlike form after 25 fs, which is elongated in the direction of motion by only about 30% compared to the initial size of C_{60}. On the other hand, the Yukawa potential would give in this case an exploded sphere of about 2.1 nm in diameter after 25 fs, which is 3 times the equilibrium diameter of C_{60}.

These rather counterintuitive results for Coulomb explosions of fast C_{60} clusters in solid targets show that the wake forces of the electron gas may stabilize the cluster, or at least a good part of it, against the explosion. This is certainly good news for applications of fast cluster-ion beams where large energy deposition in the target is desired for prolonged times. The above mentioned reduction of ion charges due to the vicinity effect in the cluster [20] may contribute only to further taming of the Coulomb explosion. Particularly intriguing are scenarios, like the one shown in Fig. 2, where the leading part of the cluster expands, while the trailing part is compressed by the wake forces, and one may speculate about an arising precursor for a wake-regulated partial fragmentation of the cluster in the target. Moreover, the role that the wake forces play in Coulomb explosion strongly depends on cluster speed, which may be then used as a tuning parameter, together with the size of the cluster, for achieving a variety of explosion scenarios, depending on the desired application of the cluster.

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*Email address: ynwang@dlut.edu.cn

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