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Fragmentation and Coulomb explosion of deuterium clusters by the interaction with intense laser pulses

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Experiments of Zweiback *et al.* [Phys. Rev. Lett. **84**, 2634 (2000)] on the interaction of intense femtosecond laser pulses with a dense molecular beam of large deuterium clusters have shown that these clusters can lose most of their electrons and explode, in a process known as Coulomb explosion. The collisions between the fast deuterium (D) nuclei give rise to D-D fusion. This has motivated us to carry out computer simulations based on the time-dependent density-functional theory in order to understand the ultrafast processes occurring under these high excitations. In particular we have studied the laser irradiation of the singly charged cluster D_{13}^{+}. The simulations show the occurrence of two different cluster fragmentation behaviors, depending on the intensity of the laser pulse: For not too large intensities, the cluster becomes disassembled in a slow way, whereas for large laser intensities substantial ionization takes place and a violent explosion occurs due to the electrostatic repulsion between the nuclei following the loss of the electrons by the cluster. The fast fragmentation mode fits well into the idea of the Coulomb explosion.

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I. INTRODUCTION

The study of the interaction between matter and intense laser fields (intensities higher than 10^{14} W/cm^2) with ultrashort pulse duration ($\tau < 10^{-13}$ s) has been an area of growing activity over the last two decades. The understanding of the dynamical aspects of these phenomena has progressed at the same time as the technology of ultrashort pulsed lasers to such an extent that femtosecond lasers are now widely available. The early observation of new processes, e.g., above-threshold ionization [1] and high-order-harmonic generation [2], or the more recent experimental observation of a new decay channel of excited molecules termed interatomic Coulombic decay [3], has stimulated a great number of experimental and theoretical works.

A detailed theoretical understanding has now been established for both atoms and molecules. But, as a form of matter intermediate between molecules and bulk solids, atomic and molecular clusters have attracted much attention [4,5] because they exhibit unique properties. Hydrogen clusters are a typical example of molecular clusters; with a high intramolecular binding energy of 4.8 eV and a very weak binding between the different molecules in the cluster, much research is being carried out on them. Charged clusters such as $H_3^{+}(H_2)_n$, with $n=1,2,\ldots$ formed by a trimer ion H_3^{+} solvated by neutral molecules forming one or more shells around the charged trimer, play an important role in the stratosphere and in the chemistry of interstellar clouds [6], and attention has focused on them already in the past [7,8] due to the long-range attractive forces that dominate ion-molecule interactions. The quantum nature of large hydrogen clusters has been another area of interest [9,10]. The caloric curve of size-selected $H_3^{+}(H_2)_n$ clusters has been determined in high-energy collision experiments and has been interpreted as indicating the transition from a bound cluster to the gas phase [11,12].

The irradiation of a dense molecular beam of deuterium clusters by an intense femtosecond laser has been able to drive nuclear fusion reactions [13,14]. Laser irradiation leads to the multiple ionization of the clusters, which then explode under the action of the repulsive Coulomb forces between the nuclei of the ionized atoms. Using intense and fast enough laser pulses (35 fs pulses were used in the experiments) the deuterium clusters are stripped of most of their electrons. This maximizes the Coulomb energy that is released into kinetic energy of the exploding ions. These nuclei can then collide with nuclei ejected from other clusters in the plasma. If the kinetic energies are high enough (greater than a few keV), D-D nuclear fusion driven by Coulomb explosion (NFDCE) events can occur with high probability. For large deuterium clusters the nuclei ejected have kinetic energies of many keV. Measurable neutron yields start at cluster radii of about 47 a.u., and rapidly increase by more than an order of magnitude. Apart from the obvious interest in order to fuel future thermonuclear devices, this technique may lead to the development of table-top neutron sources [13]. As the reaction that rules the process is $D+D \rightarrow {}^{3}He+n$, the neutron ($n$), released with an energy of 2.45 MeV, could potentially be used for a vast array of applications in neutron radiography [15] or materials research [16].

Motivated by those experimental works, theoretical studies of small clusters of the $H_3^{+}(H_2)_n$ family have been performed at various levels of theory, such as Hartree-Fock and Möller-Plesset perturbation theory [17,18], coupled-cluster [19], density-functional theory (DFT) [20–22], and configuration-interaction methods [23]. These theoretical works are mainly dedicated to describing the static properties of those odd-sized H_n^{+} cluster ions, especially the ground-state electronic and atomic structures, isomeric structures, and vibrational frequencies, and to the discovery of particularly stable sizes.

The aim of the present paper is to describe the dynamical process of the fragmentation of clusters irradiated by an intense femtosecond laser. In particular we study the irradiation of the singly charged cluster D_{13}^{+}, through the simulation of the first stages in the fusion experiments of Zweiback *et al.* By studying the response of the cluster to laser pulses using time-dependent density-functional theory [24] for the evolution of the electronic density and classical molecular dynamics for the motion of the nuclei, the simulations display the occurrence of two different cluster fragmentation behaviors, depending on the intensity of the laser pulse. For not too large laser intensities the cluster becomes disassembled in a relatively slow way with no loss of electrons, whereas for large laser intensities a fast dissociation occurs due to the repulsion between the nuclei following the loss of several electrons by the cluster. The latter fragmentation mode fits well into the scheme of a Coulomb explosion.

II. METHOD

The computational scheme we have used in the computer simulations is a real-space, real-time implementation of the time-dependent density-functional theory (TDDFT), where the time-dependent Kohn-Sham equations giving the evolution of the Kohn-Sham orbitals are explicitly integrated. A full description of the state of the art in this theory can be found elsewhere [24], so we briefly comment here on only the main points. The ground-state electronic structure of the initial field-free cluster is calculated using the DFT formalism; then this ground state is perturbed by a classically treated laser pulse and the time-dependent Kohn-Sham equations are propagated in real time. For the exchange-correlation energy functional we have used the adiabatic local density approximation with the Perdew-Wang parametrization [25]. The calculations have been performed with OCTOPUS [26], a recently developed code for TDDFT calculations. The simulation cell is a sphere with a radius of 15 or 17 a.u. In this volume a Cartesian grid with a constant mesh spacing of 0.4 a.u. is used. A time step of 0.1 a.u. has been chosen in the time propagation of the one-electron wave functions, with a total integration time between 1000 and 2000 a.u., depending on the peculiarities of each simulation (1 a.u. is equal to 0.0242 fs). The algorithms used to propagate in time the Kohn-Sham equations are the approximated enforced time-reversal symmetry (AETRS) algorithm to approximate the evolution operator $U(t+\Delta t,t)$, and the Lanczos subspace approximation to approximate the exponential of a matrix operator [27].

III. STATIC STRUCTURE OF THE D_{13}^{+} CLUSTER

In the above-mentioned experiments of Zweiback and coworkers [13,14] the size of the deuterium clusters is between 10^2 and 10^6 atoms. These sizes are too large, so for our TDDFT calculations we have selected the smaller cationic cluster D_{13}^{+}, since we expect that the qualitative picture of the fragmentation may not be too sensitive to the cluster size. The optimized ground-state geometry and the corresponding electron density are shown in Fig. 1(a). We can see in the center of the cluster a trimer D_3 with the form of a near equilateral triangle with two sides of length 1.87 a.u. and one of length 1.81 a.u. This triangle is surrounded by a first solvation shell formed by three almost parallel D_2 molecules with bond lengths of 1.45 a.u. The axes of those molecules are oriented nearly perpendicular to the plane of the central trimer and their centers of mass are in the plane of the trimer. The remaining two molecules form a second shell at a larger distance and sit above the plane of the trimer. The bond lengths of those two molecules, 1.44 a.u., are slightly shorter than those of the first shell, and closer to the bond length of a free molecule. Other relevant geometrical parameters are the distances from the center O of the central trimer to the centers of the molecules of the first and second shells (A and B, respectively). The mean values for those distances are 4.31 a.u. for O-A and 4.67 a.u. for O-B. The charge deficit of the D_{13}^{+} cluster is concentrated on the central trimer, so the notation $D_3^{+}(D_2)_5$ can be properly used for the D_{13}^{+} cluster. In fact the formation of the trimer is due precisely to this charge deficit. The electron density contours in a plane containing the central trimer are shown in Fig. 1(b). Apart from the typical density contours for the molecules and the trimer, density contours indicating the weak bonding between the trimer and the three molecules of the first solvation shell can be appreciated. The interatomic and intermolecular distances are consistent with those obtained by Stich *et al.* [22] for similar $H_3^{+}(H_2)_n$ clusters using DFT as well. However, if we compare these values with those obtained for $H_3^{+}(H_2)_5$ by Chermette and Ymmud [21], who used a hybrid DFT method including a part of the Hartree-Fock exchange, we notice that

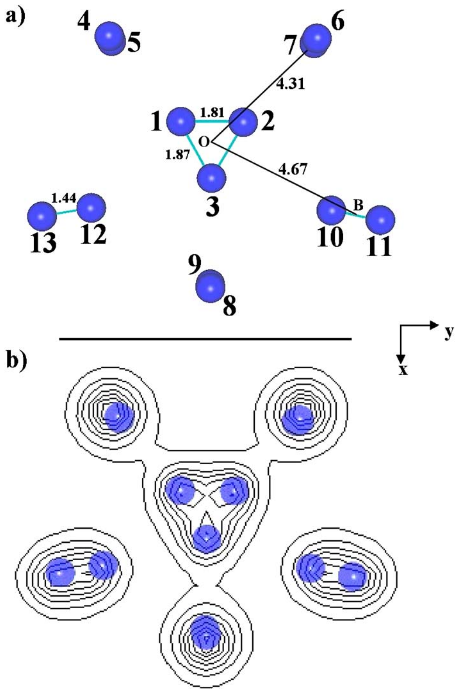

FIG. 1. (Color online) (a) Calculated lowest-energy structure of the D_{13}^{+} cluster. This structure can be interpreted as $D_3^{+}(D_2)_5$, a charged trimer solvated by five deuterium molecules forming two shells at distances of 4.31 and 4.67 a.u., respectively. Bond lengths are explicitly indicated in a.u. (b) Electron density in a plane containing the three nuclei of the trimer. Density contours go from 0.02 to 0.23 $e/\text{a.u.}^3$ in steps of 0.03 $e/\text{a.u.}^3$

the bond lengths in the central trimer are about 9% higher in our case. For the bond lengths of the molecules of the first and second shells we find that our results are greater as well by 3% and 3.1%, respectively. The first solvation shell is farther from the center of the cluster by 5.3% in our calculation, and the second shell is closer by 13.8%. So the global structure of the cluster in its ground state is the same in both cases, but in our work the distance between the first and second solvation shells is less. In fact, the same global structure is predicted by the DFT, hybrid [21], and non-DFT calculations [17,18], and through the analysis of the vibrational frequencies of the normal modes it is known that this is an intrinsically stable cluster [17,18,21,23]. The hybrid DFT method of Chermette and Ymmud predicts nearly constant bond lengths (1.67–1.69 a.u.) in the central H_3^+ unit, independently of n for $H_3^+(H_2)_n$ with n between 3 and 9, but other *ab initio* methods predict some variation with n (see data collected in Table 1 of Ref. [19], and some of those bond lengths are more similar to our values. The bond lengths of the hydrogen molecules surrounding the central trimer core obtained by other *ab initio* methods (see data collected in the previously cited table) are intermediate between our results and those of Chermette and Ymmud. We are not aware of experimental data for these clusters but we can notice that all the methods discussed above give bond lengths of the solvating hydrogen molecules very similar to the experimental bond length of free H_2, which is 1.40 a.u. The small differences between the intramolecular and intermolecular bond lengths are irrelevant for the problem that we discuss in the following sections.

IV. FRAGMENTATION OF THE CLUSTER

The main objective of this work is the theoretical study of the fragmentation of the cluster D_{13}^+ induced by the interaction with intense laser radiation. The initial purpose was to simulate the Coulomb explosion process. The available literature does not provide exhaustive information with regard to the parameters of the laser pulses, so we had to carry out a great number of simulations to find the proper parameters. The frequency of the laser radiation is an important parameter in the experiment, in order to achieve an efficient coupling of the laser light with the cluster medium leading to the largest absorption possible [28]. Consequently, we first performed a calculation of the excitation spectrum of the cluster in order to identify large absorption frequencies. The first large peak in the absorption spectrum was found at a frequency $\omega = 0.352$ a.u. (this corresponds to an energy $\hbar\omega = 9.58$ eV). Having fixed this parameter we then performed many simulations modifying the amplitude (that is, the intensity) of the laser pulse and we report here two especially interesting cases.

A. Slow fragmentation

First we present results of a simulation in which a 9.6 fs laser pulse is applied to the cluster and the dynamical evolution is followed by propagating the system for 20 000 time steps according to the time dependent Kohn-Sham equations, which are integrated with a time step of 0.1 a.u.; this gives a total simulation time of 2000 a.u. (about 48 fs). The laser pulse has a frequency $\omega = 0.352$ a.u., which is a resonance frequency of the absorption spectrum, and its shape is given by a cosinoidal envelope with an amplitude of 0.02 a.u., giving a total pulse intensity of 1.4038×10^{13} W cm^{-2}. The simulation is performed in a spherical cell with a radius of 15 a.u. The results of the evolution of the cluster structure are presented in Fig. 2, where we have selected a few snapshots for increasing time. The main features of this process are clearly seen. The central trimer absorbs energy from the laser pulse and splits into two fragments: the atoms labeled 1 and 2 in Fig. 1 form a molecule which is emitted upward (in the orientation used in Figs. 1 and 2), whereas the atom labeled 3 moves opposite to the direction of motion of the molecule. As the 1-2 molecule moves upward it passes so near to the two molecules of the first solvation shell formed by atoms 4-5 and 6-7 that they experience the intermolecular repulsive wall, and the latter two molecules are also set in motion. On the other hand, atom 3 moves down toward the 8-9 dimer. The dimers 10-11 and 12-13 of the second solvation shell appear not to be seriously perturbed in this process.

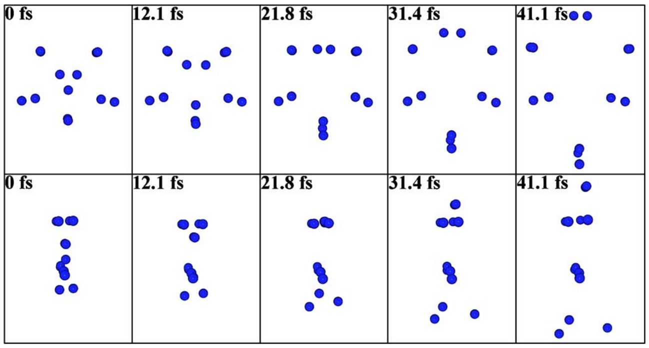

FIG. 2. (Color online) Snapshots of the cluster structure at different times after application of the laser pulse in the slow fragmentation case. Two mutually perpendicular views are presented for each snapshot.

Representing the distance of each atom to the center of the cell in Fig. 3 we can observe the following steps. The cluster roughly maintains its structure during the time of the

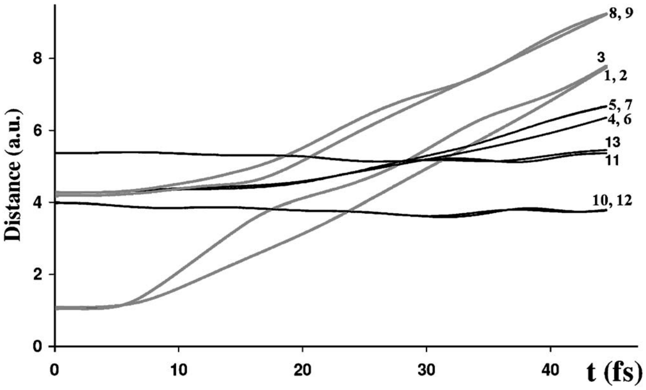

FIG. 3. Distance from each atom to the center of the cell, as a function of time. The numbers on the right side of the lines correspond to the label of each atom as seen in Fig. 1(a).

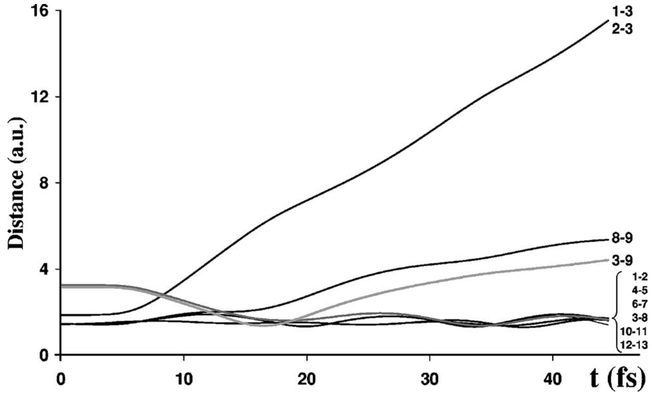

FIG. 4. Distances between some atoms as the dissociation proceeds. Notice, in particular the formation of the molecule by the atoms labeled 3 and 8 after the collision of atom 3 with the 8-9 molecule.

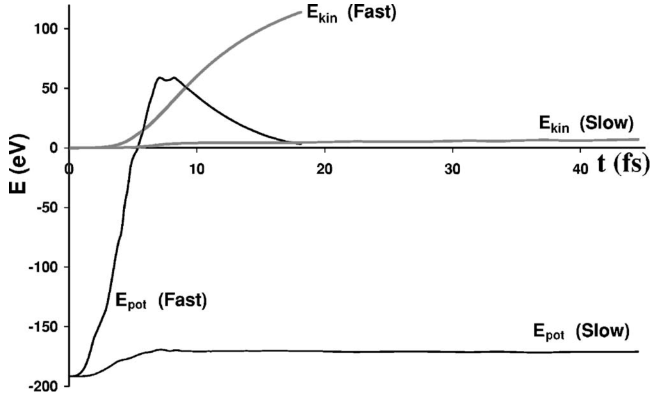

FIG. 5. Potential energy and kinetic energy of the cluster as a function of time for the cases of slow fragmentation and fast fragmentation (Coulomb explosion). Notice the Coulomb barrier in the second case.

applied pulse (the initial 9.6 fs of the simulation), due to the inertia of the atoms and the time the central trimer needs to absorb the necessary energy to break the intramolecular bonds. The only noticeable event in this initial interval is the fragmentation of the central trimer, which begins at $t=5$ fs. The trimer is split and the extra energy absorbed from the laser is transformed into kinetic energy of the two fragments, which fly in opposite directions with velocities inversely proportional to their masses. When atom 3 approaches the 8-9 molecule, an exchange collision occurs: a new molecule is formed by the atoms 3 and 8, whereas atom 9 is ejected. This can be clearly seen in Fig. 4, which shows the distances between the atoms as the fragmentation of the cluster proceeds. When the molecule formed by atoms 1 and 2 passes near the molecules 4-5 and 6-7, these molecules fly away, but each of these molecules remains bound. In Fig. 4 we see that the bond lengths of the 4-5 and 6-7 molecules simply oscillate about the equilibrium value. On the other hand, as the cluster dissociates, the D_2 molecules of the second solvation shell, that is, the molecules formed by atoms 10-11 and 12-13, are not much affected. Neither their distances to the initial center of the cluster nor their bond lengths are modified in a significant way. The molecules simply experience weak internal vibrations.

Summarizing, what we have observed here is that under the applied laser conditions, the cluster dissociates due to the initial fragmentation of the central trimer into a D_2 molecule and a deuterium atom. We have also observed the interesting process of an exchange collision between the deuterium atom and a molecule of the first solvation shell. In that collision the molecule is first dissociated and then reformed, and an atom is exchanged in the process. The fragmentation of the cluster leads then to six molecules D_2 and a single D atom flying away.

Before irradiation all the cluster energy is obviously potential energy stored in the intra- and intermolecular bonds (negative potential energy). When the laser pulse is applied, the potential energy increases (that is, it decreases in absolute value) and the kinetic energy also increases, as one can appreciate in Fig. 5. The energy absorbed by the cluster from the pulse is 28 eV. A substantial fraction of that energy, 21 eV, can be considered as potential energy, and a smaller fraction of the absorbed energy, 7 eV, is transformed into kinetic energy of the molecules of the fragmenting cluster. The change in potential energy begins almost immediately whereas the change in the kinetic energy of the molecules is delayed by about 3 fs. The laser energy is first absorbed in the form of electronic excitations of the cluster. A part of this excitation energy is employed in dissociating the central trimer and in breaking the weak bonds between the trimer and the surrounding molecules, and another part is transformed into kinetic energy of the molecules; but this does not exhaust the absorbed energy and some energy still remains as electronic excitations during the simulation. In spite of the change of potential energy observed in Fig. 5 the negative potential energy remains very substantial after the dissociation because the system still maintains most of its initial binding energy in the intramolecular bonds.

The fragmentation of the cluster begins with the dissociation of the central trimer. This is the most important step in the process, since the subsequent interaction of the products with the solvating molecules leads to the fragmentation of the whole cluster. The breaking of the central trimer is due to its efficient absorption of the energy of the laser pulse. The absorbed energy is sufficient to break the intramolecular bonds between atom 3 and atoms 1 and 2 and to impart some kinetic energy to the dissociated fragments, the 1-2 molecule and the atom 3. The analysis of the electron density distribution indicates that the initial charge deficit of D_3^+ is carried by the dimer after dissociation, and not by atom 3. This may appear surprising at first sight because the energy required for the reaction

$$D_3^+ \rightarrow D_2^+ + D \tag{1}$$

is higher than the energy for the alternative reaction

$$D_3^+ \rightarrow D_2 + D^+ \tag{2}$$

by an amount of 1.83 eV. The reason for this difference lies in the different ionization potentials of D_2 (15.46 eV) and D (13.6 eV). Nevertheless the laser pulse provides enough energy for either of the two reactions to occur; in fact the pulse

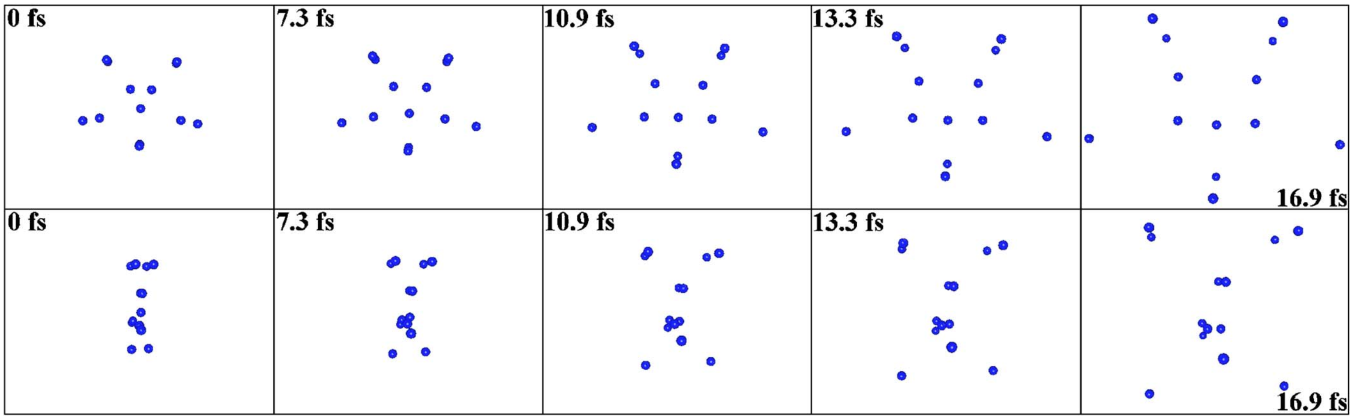

FIG. 6. (Color online) Snapshots of the evolution of the cluster structure after application of the laser pulse in the fast fragmentation case. Two mutually perpendicular views are presented for each snapshot.

provides extra energy even to set the dissociated fragments in motion.

In a second step the dissociation of the central trimer causes, as discussed above, the breaking of the weak intermolecular bonds that maintained the cluster stable. In the exchange collision between the single deuterium atom (atom 3) and the 8-9 dimer, one could have expected the formation of a trimer if atom 3 had been a D^+ cation but, since that atom is not charged, an exchange collision occurs instead. The two molecules of the outer solvation shell remain nearly unperturbed during the cluster dissociation: neither their original positions nor their internal bond lengths change. This behavior occurs because the distances of these two molecules to the cluster core, that is, to the region where the dissociation reaction starts, are relatively large. Also, their electronic coupling to the species directly involved in the reactions, that is the wave function overlap, is small, as one can observe in Fig. 1(b).

We have explored too the influence of the laser frequency. For this purpose we have performed simulations with pulses of the same amplitude as above, 0.02 a.u., but half the resonance frequency, that is, $\omega=0.176$ a.u. In this case the cluster is transparent to the laser. The atoms simply oscillate around their equilibrium positions, but the cluster does not dissociate.

B. Fast fragmentation

When the amplitude of the laser pulse is substantially increased compared to the pulse leading to slow fragmentation, while the other parameters of the pulse remain similar, the outcome of the simulation changes drastically. We have performed simulations with pulses of frequency $\omega=0.352$ a.u. and a shape given by a cosinoidal envelope with an amplitude of 0.1 a.u., five times larger than the one used in the slow fragmentation case. The duration of the pulse is again 9.6 fs, giving rise to a total intensity of 3.5095×10^{14} W cm^{-2}. The time-step used in the simulation is 0.1 a.u., as before, but since the fragmentation is faster in this case, it is enough to propagate the Kohn-Sham wave functions for 10 000 time steps; that is, a total integration time of 1000 a.u. A radius of 17 a.u. is taken for the simulation cell. The results of the evolution in time of the cluster structure are presented in Fig. 6, where a few snapshots have been selected. The process of cluster fragmentation is very different from the slow fragmentation discussed above. The main feature is that not only does the central trimer dissociate, but the intramolecular bonds of the other molecules are broken too. The atoms are then ejected as in an explosion. This sudden explosion occurs simultaneously in all parts of the cluster.

From the distances between the two atoms of each of the original molecules, shown in Fig. 7, it is clear that all the molecules dissociate (including the trimer), as the atoms repel each other as a consequence of the massive ionization; this point is discussed below. One can deduce from the individual velocities in Fig. 8 that all the atoms begin to move at about the same time, but the motion of the atoms in the outer region of the cluster is much faster (atoms 4, 5, 6, 7, 8, 9, 11, and 13). Somewhat peculiar behavior of atoms 10 and 12, and to some extent also atom 3, can be noticed in Figs. 8 and 9. These three atoms stay relatively near to their original positions. The reason is that, as the atoms 10 and 12 fly apart from their partners in the molecules (atoms 11 and 13, respectively) and atom 3 flies away from atoms 1 and 2, those three atoms 3, 10, and 12 move toward the same region of

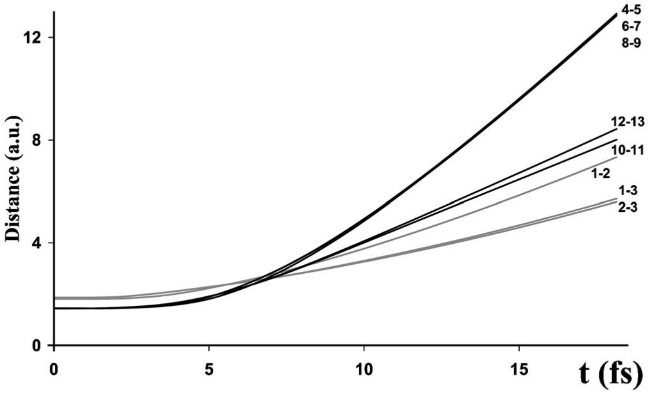

FIG. 7. Distances between the two atoms of the original molecules (and between the atoms of the central trimer) as the dissociation proceeds in the fast dissociation case.

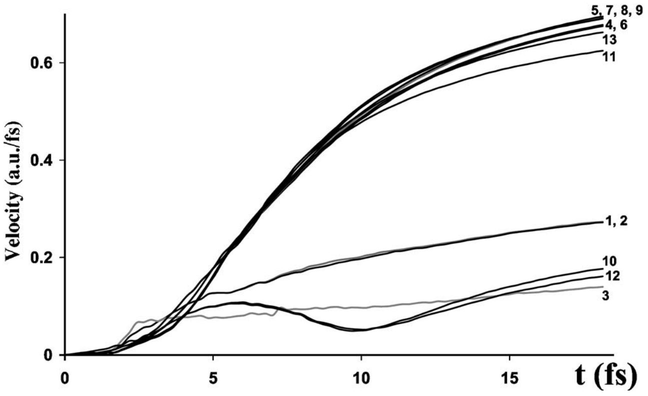

FIG. 8. Velocities of the different atoms with respect to the center of the cell in the fast dissociation case.

space; since they are ionized (see below), their mutual Coulomb repulsion slows down their motion. This is evident from the small velocities in Fig. 8.

The potential energy given in Fig. 5 quickly increases (decreases in absolute value) when the cluster interacts with the laser pulse. This indicates a loss of binding energy, and is a manifestation of the dissociation of the trimer and all the D_2 molecules. The potential energy even becomes positive (repulsive interaction) until it reaches a maximum at 7–8 fs. This repulsive potential energy arises from the electrostatic repulsion between the ionized atoms. A Coulomb explosion of the cluster then occurs and as the positively charged nuclei fly away the positive potential energy decreases showing the typical r^{-1} behavior of the Coulomb interaction. During the explosion the repulsive potential energy accumulated at the top of the Coulomb barrier is transformed into kinetic energy of the ionized atoms.

Coulomb explosion is a violent dissociation process that occurs in molecules or clusters when they are multiply ionized by laser pulses of very short duration [29]. This multiple ionization produces unbalanced positive charges sufficiently close together to cause a repulsion-induced explosion. The potential energy stored in the system due to the Coulomb repulsion between the positively charged nuclei is rapidly released as kinetic energy shared between the nuclei that fly apart. In small clusters, stripping two electrons may be sufficient to cause a Coulomb explosion [18]. In fact, cluster size seems to be an important parameter to determine whether the cluster will follow this decay channel [30]. This is the mechanism corresponding to the fast dissociation process displayed in Fig. 6. This becomes clear also from the information given in Fig. 10, where we have represented the number of electrons in the cluster as the simulation progresses. The number of electrons in the cluster is identified with the total electronic charge inside the simulation cell. In a short interval, the first 9 fs, roughly corresponding to the duration of the laser pulse, the cluster loses about five electrons. Moreover the loss of electrons does not stop at the end of that interval and it continues at a slightly slower rate until the end of the simulation without a sign of stopping.

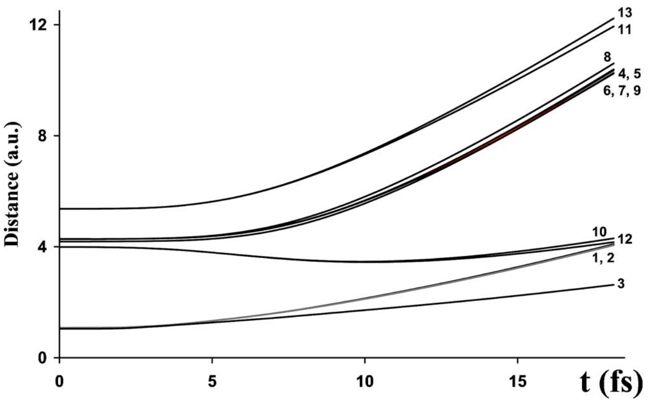

FIG. 9. Distance from each atom to the center of the cell as a function of time, in the fast dissociation case.

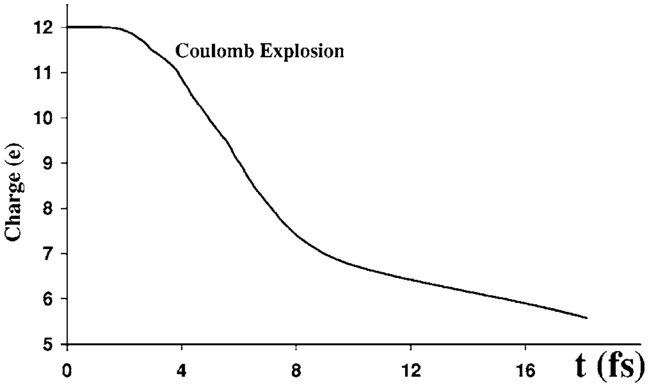

FIG. 10. Number of electrons in the cluster as the simulation progresses. This number is obtained by integrating the electron density in the volume of the simulation cell.

As a consequence of the fast ionization the cluster becomes highly charged, the electric potential energy rapidly increases, reaching a highly unstable situation, and the cluster blows up. The beginning of the Coulomb explosion, at about 8 fs, roughly coincides with the change in slope of the curve giving the electronic charge lost by the cluster in Fig. 10. When the molecules have lost their electrons, two kinds of Coulomb repulsion occur: intramolecular Coulomb repulsion and repulsion between nuclei of different molecules. The first one is stronger because the two atoms in the molecule are at a distance of 1.44–1.46 a.u., substantially shorter than the intermolecular distances.

There is, besides the size of the cluster, another key parameter in a Coulomb explosion process. If the loss of electrons is fast enough, the Coulomb repulsion drives the ions to the outer part of the cluster with substantial kinetic energy [30]. But if the loss of electrons is not fast enough, the cluster does not undergo pure Coulomb repulsion as it carries out a hydrodynamic expansion which is not as energetic as a Coulomb explosion, and of course is not energetic enough to produce the D-D nuclear fusion observed by Zweiback and co-workers [14]. The cluster must be stripped of almost all their electrons in a time scale of a few femtoseconds for a pure Coulomb explosion to occur. Laser-based techniques have so far not been so effective since the time they took to strip many electrons was typically longer [31], but in recent years very-high-power sub-10 fs pulses have become available, greatly improving the situation [32].

We can notice that all the conditions characterizing a Coulomb explosion process are present in our simulation. The process observed is close to a pure Coulomb explosion, although one can notice an incipient dissociation before reaching the maximum of the repulsive barrier.

The velocity of the fastest nuclei resulting from this Coulomb explosion (those of the outer shell) is about 0.66 a.u./fs, which corresponds to a kinetic energy of 0.013 keV. Nuclear fusion is not at all an easy process because of the substantial Coulomb barrier between the reacting nuclei. But, once the barrier is overcome, fusion becomes very likely as the two overlapping nuclei quickly reach a state of minimum energy. In most fusion processes, from controlled fusion reactors to solar reactions, the reacting particles have energies in the range of 1–10 keV [33]. This indicates that the Coulomb explosion process simulated in this work delivers kinetic energies that are still far from those required to produce D-D fusion. This is an expected result since the radius of the D_{13}^{+} cluster is only 5 a.u., whereas the clusters in the experiments of Zweiback *et al.* are much larger; their radii are ten times bigger, so the average number of atoms is 1000 times larger. The initial radius R of the cluster determines the maximum energy produced in the explosion [14]

$$E_{\max} = \frac{q n_{\mathrm{D}} e^2 R^2}{3\epsilon_0} \qquad (3)$$

where q is the charge state of the cluster and n_{D} is the initial density of deuterons. The maximum energy is then proportional to R^2 and a deuterium cluster with a radius greater than 50 a.u. will be necessary to produce the multi-keV ions required for NFDCE [14].

Last and Jortner [34,35] have recently proposed, and shown by molecular dynamics simulations, that very energetic deuterium or tritium nuclei (D^+ or T^+) can be produced by the multielectron ionization and Coulomb explosion of molecular heteronuclear clusters of D on T bound to heavy atoms, e.g., clusters formed by D_2O and T_2O molecules, similar to water clusters. These clusters will provide substantially higher fusion reaction yields than homonuclear clusters of the same size.

V. CONCLUSIONS

We have performed computer simulations of the fragmentation of the cluster $D_3^{+}(D_2)_5$ induced by the interaction with an intense, ultrafast laser pulse, using time-dependent density-functional theory. The analysis of the results reveals two different decay channels. In the first one, the fragmentation process is relatively slow and it occurs sequentially: the laser pulse induces the dissociation of the central trimer and, in a second step, the motion of the dissociated fragments induces the destabilization and dissociation of the cluster. The second dissociation channel, observed for laser intensities about 20 times larger compared to the slow fragmentation case, is a fast and violent process known as Coulomb explosion. The cluster suffers substantial ionization and the bare nuclei are accelerated due to the repulsive Coulomb forces and ejected from the cluster with high kinetic energies. As the TDDFT is able to simulate efficiently a Coulombic explosion process in small deuterium clusters, we plan to study much larger clusters, in order to make closer contact with the experiments in which Zweiback *et al.* [14] observed the occurrence of nuclear fusion reactions with production of neutrons.

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