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Simulation of exploding clusters ionized by high-intensity femtosecond laser pulses

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The dynamics of small ($\leqslant$55 atom) argon clusters ionized by a high-intensity, femtosecond laser pulse have been studied using classical particle dynamics simulations. The evolution of both ions and electrons during the laser pulse is examined. These simulations show that the space-charge and impact ionization of electrons, liberated from individual atoms in the cluster, have a very important effect on the ion energies resulting from the cluster explosion. [S1050-2947(98)50106-0]

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While the multiphoton ionization of van der Waals clusters with intense nanosecond laser pulses has been studied for many years [1,2], only recently has the ionization and fragmentation of atomic clusters by very-high-intensity, ultrashort light pulses received substantial experimental [3–12] and theoretical [6,13,14] attention. In particular, studies of the energies of ions produced in the explosion of clusters ionized by intense laser pulses have shown some remarkable results. Purnell *et al.* reported on the energies and charge states of ions produced in the multiphoton ionization of small HI clusters and co-clusters of Ar and HI with 350-fs, 624-nm laser pulses at intensities up to 10^{15} W/cm^2 [4]. They observed I and Ar ions with charges as high as 8^+ and energies of up to 500–1000 eV, which they attributed to a Coulomb explosion of a highly charged cluster created by multiphoton ionization. Last *et al.* have shown with molecular-dynamics simulations that these observed ion energies were not consistent with vertical ionization of the cluster prior to any ion motion [14].

Recently, experimental studies of the intense laser ionization of Xe clusters by Ditmire *et al.* indicated that the Coulomb explosion picture, however, is not entirely accurate for larger clusters [10,11]. In Ref. [10] Xe clusters with up to 2500 atoms were irradiated with 780-nm, 100-fs laser pulses at intensities up to 2×10^{16} W/cm^2. These experiments observed Xe ions ejected from the clusters with energies up to 1 MeV and charge states as high as 40^+. Using a plasma model for the cluster explosion it was shown that the high charge states result from electron collisional ionization of the cluster ions, and that the observed ion energy spectrum was the result of hot electrons, confined to the sphere of the highly ionized cluster by space-charge forces, driving the expansion of the cluster in a hydrodynamic manner [6,11].

In this Rapid Communication, classical particle dynamics simulations are presented that examine intense, femtosecond ionization of small to medium size argon clusters and the subsequent energetic explosion of the ions. Unlike previous calculations [14], these simulations include the motions and fields of both ions and electrons produced during ionization, as well as the effects of electron-impact ionization. These calculations indicate that even for clusters as small as six atoms, the influence of the electrons' fields must be included to accurately model the disassembly dynamics of the clusters.

To study the fragmentation dynamics of van der Waals bonded clusters ionized by a strong laser field, a classical particle dynamics including laser fields and ionization mechanisms has been developed (similar to that previously presented in Ref. [13]). In brief, the evolution of the ions in the cluster is computed using standard molecular-dynamics methods. The initial structure of the argon clusters is chosen to be that of a closed-shell icosohedron (13- or 55-atom cluster) [15], with an atom spacing of 3.7 Å [16]. Six atom clusters are given an initial octahedral structure. Because the Coulomb repulsion forces between ions dominate the cluster dynamics, they are the only forces used in the calculation. (A small number of calculations were performed in which a Lennard-Jones potential was included between neutral and singly charged ions; however, its inclusion was found to have virtually no effect on the outcome of the simulations.) To account for the finite size of the electron cloud around each ion point charge, the ion's potential is modeled as a soft-core potential $U(r)=Ze/\sqrt{r^2+a^2}$, where a is chosen to yield a potential well with the same depth as the ionization potential.

The atoms of the cluster have no initial kinetic energy and are subject to an oscillating electric and magnetic laser field. The pulse used in the simulations has a wavelength of 800 nm and a temporal full width at half maximum of 100 fs. Ionization of Ar atoms at the intensities considered here is primarily by tunnel ionization [17]. To account for this, the electric field at the position of each ion is found and the dc tunneling ionization rate W_{tun} is calculated [18]. The probability for ionization during a time step is given as $W_{\mathrm{tun}}\Delta t$. Whether a given ion is ionized during a time step is determined by a comparison of a random number to the tunneling probability. This method has the advantage of more accurately predicting tunnel ionization dynamic behavior as the laser-pulse intensity increases than the use of a simple barrier suppression model, in which an ion is immediately ionized once a threshold in the electric field is reached [19]. Upon ionization a free electron, with no initial kinetic energy, is placed near the parent ion in the direction of the ionizing field. This procedure has the effect of reproducing the above-threshold ionization electron-energy spectrum predicted by the quasiclassical model. Additional ionization through inelastic collisions of energetic electrons with ions is also included. If an electron approaches an ion with an impact parameter smaller than the collisional ionization cross-section

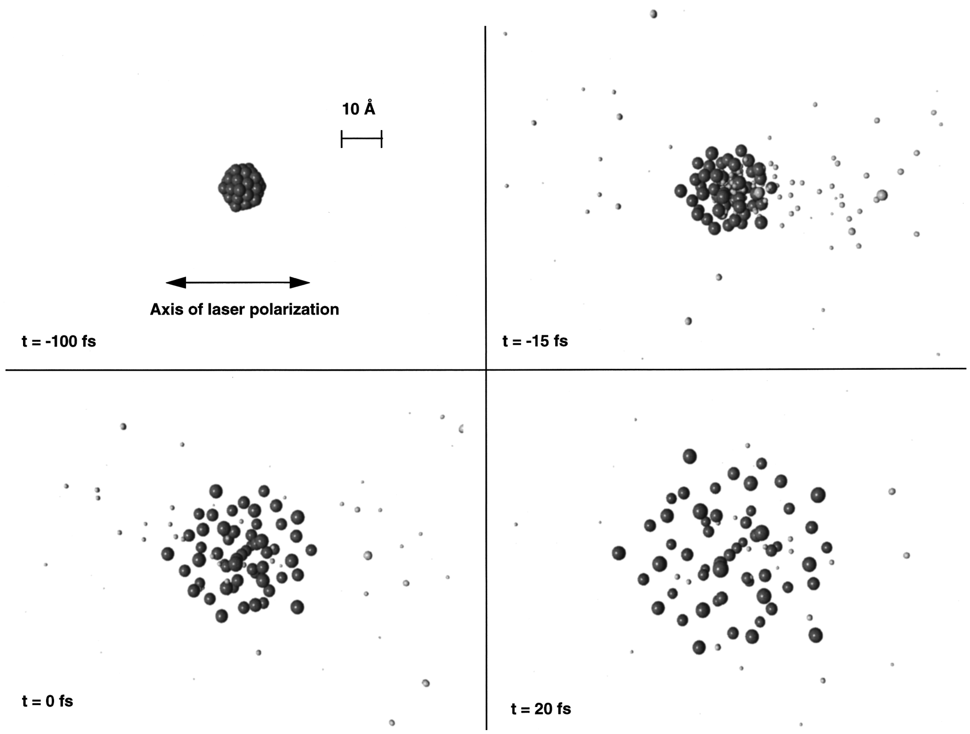

FIG. 1. Four snapshots of a 55-atom argon cluster at different times in the laser field with a peak intensity of 1×10^{15} W/cm^2. Ions are shown as the large dark particles, and the electrons are shown as the small light particles.

radius [20], the ion is immediately ionized and the new electron is given a velocity and a position near the ion so as to conserve the momentum and the total energy of the system.

The forces between the electrons and ions are found by a pairwise addition of the electric and magnetic fields in the electrostatic and magnetostatic approximations (i.e., nonrelativistic particle motion with instantaneous field propagation across the cluster). The classical equations of motion of the particles are integrated using a fifth-order, embedded Runge-Kutta-Felberg algorithm with adaptive step-size control [21]. The step size during the course of each run varied from 10^{-19} to 2×10^{-17} s, insuring better than 5% energy conservation over the course of the run. The motion of the cluster particles was propagated until the minimum separation between ions exceeded 30 Å, a value chosen since it yields better than 10% accuracy in predicting the asymptotic energy of ions ejected by mutual Coulomb forces of an initially charged cluster. Computational runs were performed on clusters as large as 55 atoms, although, because of the very large computational effort required for a cluster of this size, only a limited number of such runs have been performed. On the other hand, a large number of runs have been performed on smaller clusters, each with a different random number seed, to statistically sample the random ionization algorithm.

All of the results presented in this paper were conducted using a peak laser intensity of 1×10^{15} W/cm^2. Figure 1 shows four snapshots of a 55-atom argon cluster at different times in the laser field. (Ions are shown as the large dark particles and the electrons are shown as the small, light particles.) At a time 15 fs before the peak of the pulse (a time 60 fs after the first ionization event), the cluster has begun to expand. At this point 427 electrons have been produced by ionization and, as is clear in this image, many of them ($\sim$100 electrons) are still confined in and around the body of the cluster. The cluster itself is expanding essentially isotropically. At the peak of the pulse $(t=0)$, many of the electrons are still in the vicinity of the expanding cluster. 20 fs later the electron density in the vicinity of the cluster has fallen, and the cluster continues to expand more or less isotropically via Coulomb forces.

The history of the cluster ionization of three different cluster sizes, 6, 13, and 55 atoms, from individual runs characteristic of the dynamics, are illustrated in Fig. 2(a). Early in the pulse the ionization is dominated by tunneling ionization by the laser field. After a number of electrons have been liberated they are driven by the laser field and can continue to ionize the cluster later in the laser pulse by collisional ionization. The level of final ionization in the cluster increases as the cluster size increases. This is due to the fact that the larger space-charge forces of the larger cluster confine the electrons to the bulk of the cluster longer [6].

To examine the time history of the electrons' space-charge confinement to the cluster, the number of electrons that have exited the cluster as a function of time is plotted in Fig. 2(a) as well. This quantity is defined as the number of electrons beyond a radius of 50 Å plus the radius of the outermost ion. As can be seen in Fig. 2, in the smallest cluster of six atoms, most of the electrons exit the cluster upon ionization. During the majority of the laser interaction with the 55-atom cluster, however, a large fraction of the ionized electrons do not escape until later in the laser pulse. This is essentially due to the strong confining positive charge of the larger cluster. Nonetheless, the ionization by laser-driven electrons is important even in clusters as small as six atoms. Figure 2(b) plots the calculated average charge state reached in the 6-, 13-, and 55-atom clusters. (For the 6- and

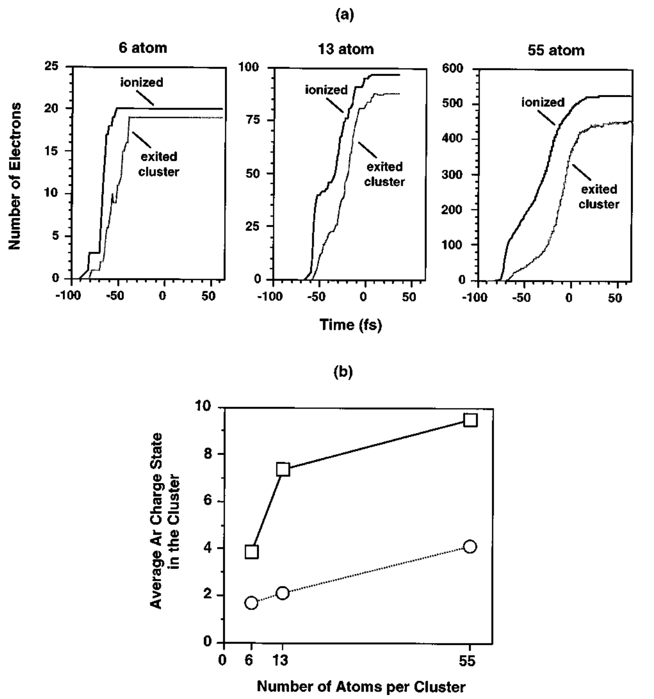

FIG. 2. (a) Plots of the number of electrons produced as a function of time in calculations involving 6-, 13-, and 55- atom Ar clusters (solid lines). The number of electrons that have exited the cluster are the dotted lines on each graph. (b) Calculated average charge state per ion produced in a cluster as a function of cluster size (squares). The same quantity calculated when electron fields and collisional ionization are ignored are also shown (circles).

13-atom clusters these average charge states were calculated as the average over ten different runs.) For comparison, the charge state reached in calculations in which laser field ionization is included, but the fields and impact ionization of electrons are ignored. For all three size clusters, the level of ionization is larger when impact ionization is included, though the difference is most pronounced in the 13- and 55-atom clusters.

The resulting ion energy spectrum of the 55-atom Ar cluster, after its explosion, is shown in Fig. 3. This figure plots a histogram of the number of ions within 290-eV energy bins. The explosion of this cluster exhibits a maximum ion energy of 2.9 keV, though the average ion energy is around 1.8 keV.

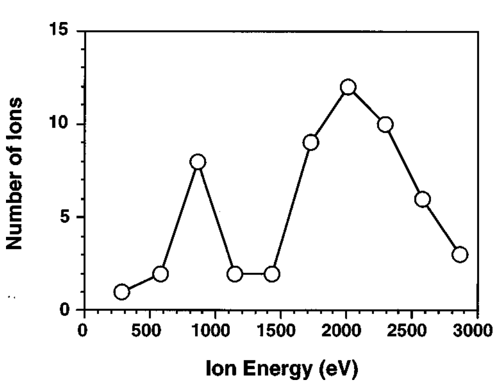

FIG. 3. Ion energy spectrum calculated in the 55-atom cluster simulation. This is a plot of a histogram of the number of ions within energy bins of a 290-eV width.

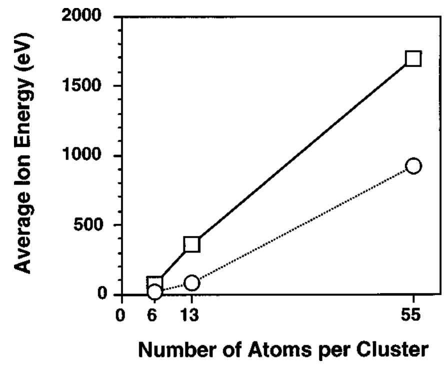

FIG. 4. Average ion energy calculated from the explosion of the clusters as a function of cluster size (squares). The average ion energy from simulations ignoring electron fields and impact ionization is also plotted here (circles).

The ion distribution exhibits two distinct peaks, a consequence of the two-shell icosohedral initial structure. The broad distribution results from two factors. The ions in the cluster are not uniformly ionized so some ions have slightly higher charge states than others. (Ions of 8^+, 9^+, and 10^+ are produced in this calculation.) Furthermore, elastic collisions of energetic laser driven electrons with the ions can broaden the energy distribution as well [11].

Figure 4 illustrates the calculated average ion energy of the 6-, 13-, and 55-atom clusters. The ion energy increases roughly linearly as the cluster size increases. A linear scaling of ion energy with cluster size is expected from a Coulomb explosion model of the cluster disassembly. For comparison, the ion energy from the explosion of the clusters calculated, ignoring electron fields and impact ionization, is also shown in Fig. 4. In general, the average ion energy is lower than in the full calculations. This is a result of the higher charge states achieved by electron collisional ionization in the full calculation. It is interesting to note that even in the case of the six-atom cluster the ion energies are higher when electron effects are included.

These calculations indicate that there appears to be a transition in the nature of high-field laser-driven explosions of clusters as the cluster size is increased. With small clusters, the classic picture of a cluster multiphoton or tunnel ionized, followed by a Coulomb explosion of the ions, appears to be largely accurate. As electrons are ionized in the small clusters, they exit the cluster volume quite rapidly. However, as the cluster size increases, the space charge of the ions confines the electrons, and they can further ionize the cluster through collisions with the ions. The transition between these two extremes is already apparent in these simulations with modest size clusters of 13 atoms or more.

This behavior has been previously predicted in larger clusters. The work of Ref. [6] found that the high charge states observed from clusters ionized by high-intensity short pulse lasers were consistent with a plasma model of the cluster interaction, which included a rate equation description of the collisional ionization. The shape of the ion energy spectra observed in the experiments of Ref. [11] also indicated that the explosion of the cluster was driven by hot electrons confined to the body of the cluster rather than by Coulomb forces alone.

It should be mentioned that these simulations may provide an explanation for the observations of Purnell *et al.* [4]. In their experiment they observed ions as highly charged as Ar^{8+} with a corresponding average ion energy of $\sim$500–1000 eV. This is not dissimilar to the charge states (7^+–8^+) and the average ion energies (400 eV) observed in the simulations presented here for the 13-atom Ar clusters, near the size of clusters used in their experiments. The fact that Purnell *et al.* only observed high charge states and ion energies when HI was seeded in the Ar expansion may be due to the fact that the HI was seen to aid the formation of larger clusters [4]. As the simulations indicate, larger clusters are more likely to exhibit the anomalously high charge states and consequent high ion energies created by electron-driven impact ionization. In addition, these simulations suggest that the hypotheses of Snyder *et al.* [8], that the ionization ignition model [13], in which laser field ionization with the aid of the fields of neighboring ions can explain the high charge states observed, is not entirely correct. The calculation presented here (Fig. 2) indicates that field ionization is inadequate in reproducing the charge states of Ref. [4]; electron collisional ionization is necessary.

In conclusion, particle-dynamics simulations of the interaction of intense, femtosecond laser pulses with small-to-medium-sized argon clusters have been presented. These simulations show that the space-charge confinement of electrons liberated by strong field ionization in larger clusters has an important consequence on the evolution of the cluster ionization and subsequent explosion. This result confirms that the vertical model of ionization cannot be accurately applied to large clusters and the model must account for the effect of the electron fields and impact ionization. Finally, these calculations are in reasonable agreement with previous experimental measurements of the disassembly of argon clusters upon ionization with an intense femtosecond laser pulse.

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