Dynamic Acceleration Effects in Explosions of Laser-Irradiated Heteronuclear Clusters

M. Hohenberger, D. R. Symes,* K. W. Madison,† A. Sumeruk, G. Dyer, A. Edens, W. Grigsby, G. Hays, M. Teichmann, and T. Ditmire

The Texas Center for High Intensity Laser Science, Department of Physics, University of Texas at Austin, Austin, Texas 78712, USA

(Received 13 September 2004; published 2 November 2005)

Intense, femtosecond irradiation of atomic and molecular clusters can initiate Coulomb explosions, generating particle energies sufficient to drive nuclear fusion. Last and Jortner have proposed, based on particle dynamics simulations, that heteronuclear clusters with a mixture of heavy and light ions will not explode by the simple, equilibrium Coulomb model but that dynamic effects can lead to a boosting of energy of the lighter ejected ions [Phys. Rev. Lett. **87**, 033401 (2001)]. We present experimental confirmation of this theoretically predicted ion energy enhancement in methane clusters.

DOI: 10.1103/PhysRevLett.95.195003 PACS numbers: 52.50.Jm, 36.40.Wa, 52.70.Nc

In recent years, there has been much activity in studying high intensity, ultrashort laser pulse interactions with atomic clusters. At high intensity ($> 10^{16}$ W/cm^2), the physics governing the laser cluster interaction is fundamentally different than in previous optical studies on clusters. Femtosecond pulses are comparable to or shorter than the disassembly times of a cluster in the laser field, and so the entire laser pulse interacts with an inertially confined body of atoms. The most dramatic consequence of this unique interaction has been the observation of high charge states and very fast ions [1–3], which can be exploited to drive nuclear fusion [4–7]. Most studies have centered on single species homonuclear clusters such as van der Waals bonded rare gas or hydrogen clusters. For large, high-Z clusters subject to intense IR, optical or vacuum ultraviolet irradiation, and moderate intensity, the space charge forces in the cluster prevent complete escape of electrons photoionized in the cluster forming a nanoplasma which exhibits a variety of collective electron oscillation effects [8,9]. In contrast, at high enough intensity, particularly in smaller, low-Z clusters, an intense, ultrashort laser pulse can ionize and expel most of the electrons from the cluster. This results in a pure Coulomb explosion, in which the ions expand nearly isotropically by mutual repulsion in the charged sphere, gaining kinetic energies simply related to their initial potential energy at their equilibrium position in the cluster [10]. If the electron extraction occurs on a time scale shorter than that for significant ionic motion, the outer ionization is termed cluster vertical ionization (CVI) [11–13]. Several models have also been developed for the intermediate regime, where a core of electrons remains bound to the cluster and can give rise to asymmetries in the explosion [14–16].

With this developing understanding of laser driven explosions in homonuclear clusters, the question exists as to whether mixed species, heteronuclear clusters behave in the same manner under intense ultrafast laser irradiation. Previous experiments have studied heteronuclear molecular clusters driven with modest intensity ($\approx 10^{15}$ W/cm^2) laser pulses [17,18] as well as HI clusters at high intensity [19] and nuclear fusion in CD_4 cluster plasmas [6,7]. The motivation for studying Coulomb explosions of mixed species clusters has been advanced through simulations conducted by Last and Jortner [11–13] that indicate two advantages of heteronuclear over homonuclear clusters. First, the energy of the light ions is boosted because the presence of the multicharged ions increases the potential driving the Coulomb explosion of the fully stripped heteronuclear cluster. This has been verified by comparing CD_4 and D_2 clusters through fusion neutron studies in Ref. [6] and ion energy measurements in Ref. [7]. Second, the acceleration of the light ions can be enhanced if they overtake heavy ions during the expansion and are thus more strongly repelled, which occurs if the kinematic parameter for species A, $\eta_A = q_A m_B / q_B m_A$, is greater than 1 (where q and m are the charge and mass of the light ion A and heavy ion B). In this case, the light ions explode in an outer shell with an average energy higher than what would be expected from the purely energetic argument based on the initial potential energy of the ions. Such a kinematic enhancement is an *isotope* effect and cannot be identified by a comparison of CD_4 and D_2 clusters as in previous studies [6,7].

In this Letter, we present an experimental confirmation of the second prediction of the theory by measuring energy spectra from laser induced explosions of the methane isotopes CH_4 and CD_4. Our laser intensity of $\approx 10^{17}$ W/cm^2 is sufficient for the clusters of radius ≈ 3 nm to be inner ionized to a charge state of $(C^{4+}A_4{}^{+})$ and for the CVI approximation to be valid [13]. As such, the cluster undergoes a symmetric Coulomb explosion as described by the Last and Jortner heteronuclear cluster model [11–13]. We observed a large difference in the energy spectra which is unambiguous evidence of a dynamic acceleration effect during the ion expansion, since the initial potential driving the cluster explosion is the same in both cases.

In our experiment, a pulsed supersonic gas jet with an orifice of $d = 750$ μm directed through a skimmer produced a low density cluster beam. The energies of the ions created by the laser cluster interaction were determined by measuring their time of flight (TOF) in a 1.14 m field free drift tube. A microchannel plate (MCP) detected ions in an angular cone of 7.8×10^{-4} sr from the focus. To estimate a mean cluster size, we employed the Hagena parameter [20] $\Gamma^* = kP_0 \cdot (d/\tan\alpha)^{0.85}/T_0^{2.29}$, where $\alpha = 5°$ is the expansion half angle, T_0 and P_0 are the reservoir temperature (295 K) and pressure (532 psi), respectively, and k is a constant related to bond formation. Assuming $k = 2360$ [21] for both CH_4 and CD_4, the cluster-size distribution is expected to be the same. These conditions predict Γ^* to be almost identical to that in a previous characterization [7] of a similar gas jet operated with CH_4, so we expect a cluster size of approximately 3 nm. To irradiate the clusters, we utilized the Texas High-Intensity Optical Research laser, which delivers 40 fs, 800 nm pulses with energy up to 0.75 J at 10 Hz. The laser was focused using an $f/4.9$ refractive graded index aspheric lens limiting the maximum energy to <0.1 J. The energy was varied between 21 and 70 mJ, providing intensities between $\approx 9.6 \times 10^{16}$ and $\approx 3.2 \times 10^{17}$ W/cm^2.

A measured ion distribution is shown in Fig. 1. The original TOF data, shown in the inset, was averaged over 200 shots, normalized, and converted to an energy spectrum using the formula $f(E) = f(t)(dE/dt)^{-1}$. The shape of the spectrum is consistent with that expected from Coulomb explosion when a distribution of cluster sizes is taken into account [5].

Figure 2 shows kinetic energy spectra at two laser intensities comparing CH_4 and CD_4 data. (The low energy components of the spectra are not shown, so the curves do not intersect.) The difference in kinematic parameters between H^+ ($\eta = 3$) and D^+ ($\eta = 1.5$) causes a clearly apparent isotope effect. The transformation to an energy

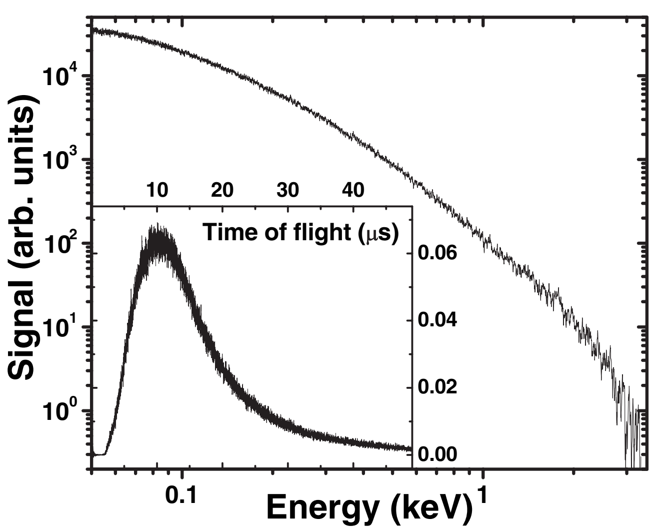

FIG. 1. Averaged energy spectrum and time of flight trace (inset) from CD_4 clusters irradiated at 2.1×10^{17} W/cm^2.

distribution required us to assume a certain mass, which we set to the mass of H^+ or D^+, according to the gas we were shooting. The signal is a superposition from both carbon and deuterium/hydrogen arriving at the MCP, and these signals cannot be separated. Our simulations on cluster explosions (see later) show, in good agreement with the results obtained in Ref. [12], that the maximum energy E_C, for carbon ions of mass m_C, is approximately 2.5–3 times the maximum energy of the lighter species. We can then estimate a minimum TOF for the heavier carbon ions which corresponds to a maximum energy in the energy spectrum obtained using the mass of the lighter ion species m_A. Any signal above this energy $E_{min} = E_C m_A/m_C$, denoted in Fig. 2 by the dotted lines, can be caused only by the detection of H^+ or D^+ ions. In the energy ranges

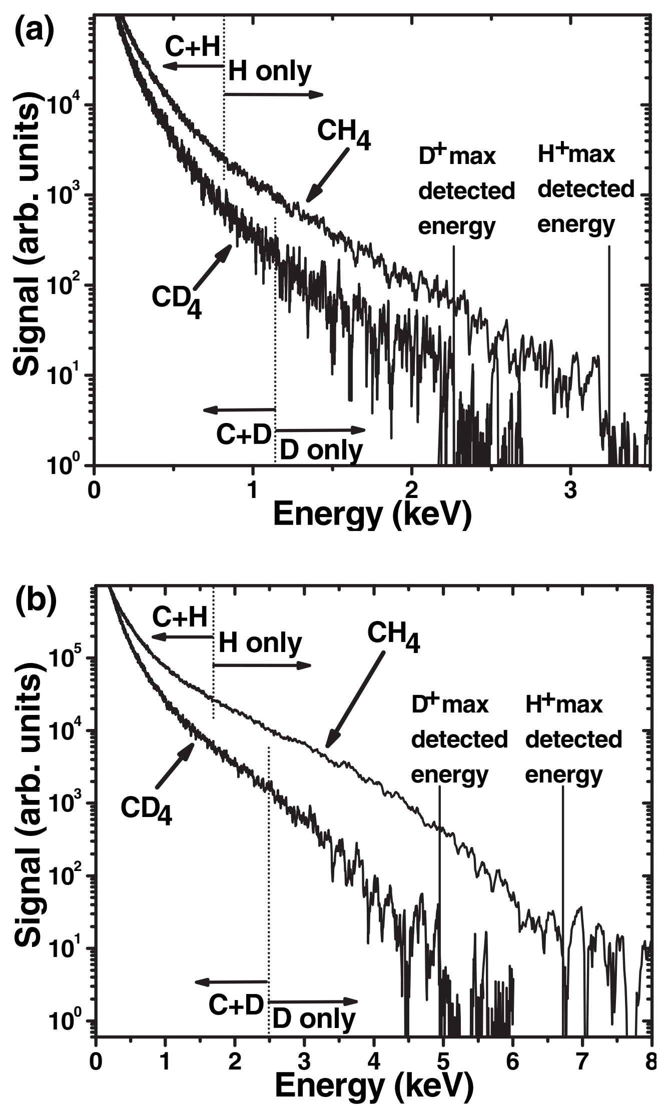

FIG. 2. Energy spectra from CH_4 and CD_4 clusters at a laser intensity of (a) 1.2×10^{17} W/cm^2 and (b) 3.2×10^{17} W/cm^2. Dotted lines indicate E_{min}, above which signals are assumed to be solely due to the light ion species.

attributed solely to the lighter ion species, the H^+ energies always exceed the D^+ energies, indicating a higher average energy in the proton distribution and validating the theory predicting kinetic energy enhancement. Although the MCP response will not be the same for H^+ and D^+, we optimized the operating parameters to minimize signal dependence on ion mass and found a signal difference of at most 10%, which is negligible compared to the observed difference in signal, which is about an order of magnitude.

This interpretation is based on the highest energy ions, which certainly originate at the most intense part of the focus where the intensity is sufficient to initiate pure Coulomb explosions and demonstrate energy enhancement. We also expect a flux of ions to be detected from cluster interactions induced in the lower intensity regions of the laser focus or from laser pedestal and prepulse. However, these explosions would have been considerably less energetic, and hydrodynamic forces would likely play a more significant role, meaning the accelerated ions would not lie in this high, multi-keV region of the energy distribution. The fact that we see an energy difference is, in itself, evidence that the ions must be driven predominantly by Coulomb forces, since, in a hydrodynamic scenario, the plasma expands at the sound speed $c_s = (Zk_BT_e/m)^{1/2}$, where Z and m are the ion charge and mass, respectively, and k_BT_e is the electron temperature. The cluster constituents would, therefore, gain the same energy $\frac{1}{2}mc_s{}^2$, in both the CH_4 and CD_4 cases.

In principle, the maximum energy is determined by the outermost particles in the cluster which never experience the dynamic energy enhancement since they do not outrun any heavy ions and so should be unaffected by the species chosen. Although our data show a difference in the maximum observed H^+ and D^+ energies, it is likely that we do not actually detect the highest energy particles, since their flux is so weak that the signal falls below the noise level. Hence, the apparent separation of maximum energy is still consistent with kinetic energy enhancement. An unexpected feature is the increase of maximum energy with laser intensity, since a pure Coulomb explosion is driven solely by ion repulsion forces, and the laser conditions beyond the saturation intensity are irrelevant. This behavior is a consequence of a cluster-size distribution, since, at the lower intensities, the larger clusters in the distribution will not be fully stripped of their electrons.

In order to help interpret the data, we wrote a simple particle simulation that models the Coulomb explosion of heteronuclear clusters. Our code is based on a 5th order Runge-Kutta algorithm with adaptive step size. All particles are treated classically. We utilize the CVI approximation [11], which assumes that on the onset of the cluster explosion ($t = 0$) all of the relevant electrons are removed from the system and do not contribute to the acting space charge forces. This allows a separation of the time scales between the removal of all unbound electrons and the spatial expansion of the cluster. We also assumed, as in Ref. [11], that the remaining bound electrons are so tightly connected to their host atoms that they do not alter the subsequent Coulomb explosion of the cluster. Because of these assumptions, both bound and unbound electrons are not explicitly treated in our model, which considerably simplifies the simulation.

Our code modeled the Coulomb explosion of methane clusters, $(CH_4)_n$ and $(CD_4)_n$, with n equals 257, 515, 925, 1419, and 2109, with the corresponding radii of 1.8, 2.2, 2.6, 3.1, and 3.5 nm, respectively. Hydrogen and deuterium were completely ionized, and the carbon was ionized to a charge state of C^{4+}. The explosion was calculated on a near picosecond time scale (0–4×10^{-13} s) starting with 10^{-17} s time steps. The clusters were modeled as consisting of uniformly distributed molecules with a C-C spacing of 4.4 Å and a C-D or C-H distance of 1.09 Å. This corresponds to a density of 1.2×10^{22} molecules/cm^3. However, the results are not expected to be sensitive to these simplifications of the cluster structure. The isotope effect is well demonstrated in Fig. 3, where the calculated energy distribution of H^+ and D^+ from a $(CA_4)_{2109}$ Coulomb explosion is shown. The distribution for hydrogen is, as expected, shifted to higher energies with respect to the deuterium. For comparison, the theoretical distribution for a homonuclear Coulomb explosion is shown in the inset (both on linear scale). Figure 4 displays the mean and maximum energy of all species as a function of molecules per cluster along with the results obtained in Ref. [12] for a $(CA_4)_{2171}$ cluster. One can see the separation of mean energy between H^+ and D^+, whereas the maximum energy shows no dependence on cluster species. Both maximum and mean energy for C^{4+} differ considerably between CH_4

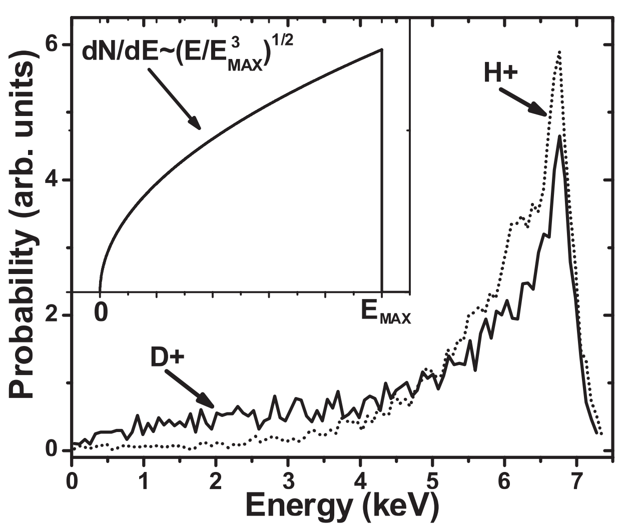

FIG. 3. Simulated energy distribution for H^+ (dotted line) and D^+ (solid line) from the Coulomb explosion of a $(C^{4+}A_4{}^+)_{2109}$ cluster. Inset: Theoretical energy distribution from a homonuclear Coulomb explosion.

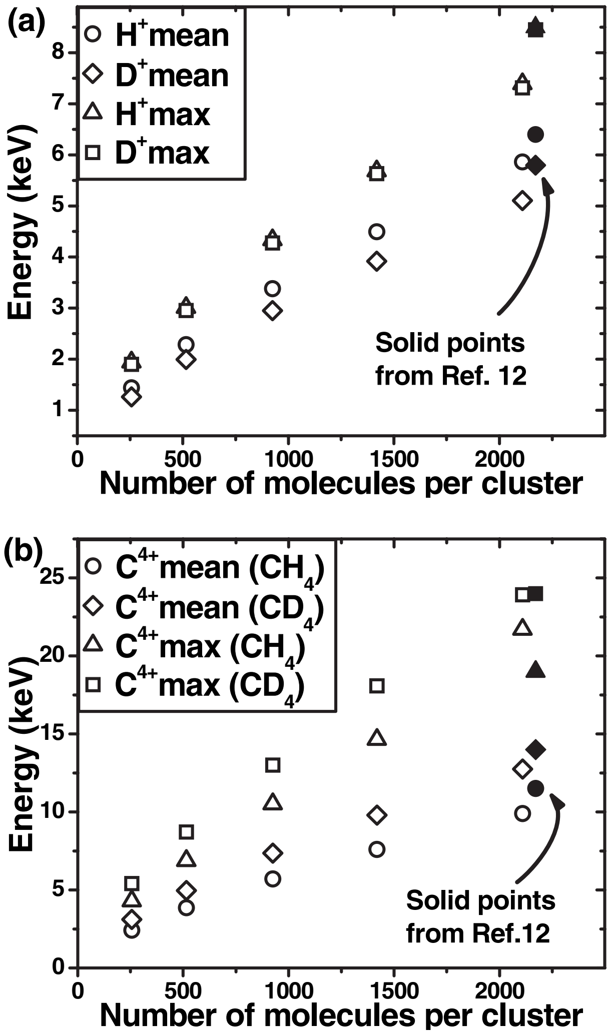

FIG. 4. Simulated mean and maximum ion energies from the Coulomb explosion of a $(C^{4+}A_4{}^{+})_{2109}$ cluster. Circles and triangles denote mean and maximum energies, respectively, from CH_4, H^+ in (a) and C^{4+} in (b). Diamonds and squares show mean and maximum energies, respectively, from CD_4, D^+ in (a) and C^{4+} in (b). The solid symbols represent the equivalent values from the simulations in Ref. [12].

and CD_4. The maximum energies of the light ions (corresponding to those on the surface) are fixed by the potential energy stored in the cluster, whereas their mean energy is enhanced as they overtake the heavier ions during the expansion. This effect is stronger for the H^+ ($\eta = 3$) compared to D^+ ($\eta = 1.5$) since it has a greater kinematic parameter. Both the maximum and the mean energies of the C^{4+} are affected by the kinematic effect as all the ions are overtaken by some fraction of the light ions which originate at smaller radii. Also, the C^{4+} from the CH_4, which has $\eta = 0.33$, suffers a greater loss of energy than the C^{4+} from the CD_4, which has $\eta = 0.67$.

In conclusion, we have clearly demonstrated the effect of kinetic energy enhancement in the Coulomb explosion of heteronuclear clusters as predicted in simulations by Last and Jortner [11–13]. We have implemented a straightforward particle simulation code which is in good agreement with this previous modeling. In methane clusters, the average kinetic energy of the resulting hydrogen ions is significantly higher than the energy of deuterium ions under identical conditions. The maximum energy of the light ions, expected to be independent of cluster species, appeared to differ between CH_4 and CD_4, although we suspect that this may be a result of detection issues at the higher end of the energy spectrum.

We acknowledge the assistance of K. Travis. This work was supported by the Department of Energy Office at Basic Energy Sciences, the Welch Foundation, Grant No. F-1501, and NNSA Contract No. DE-FC52-03NA00156.

*Corresponding author.
Electronic address: dansymes@mail.utexas.edu
†Current address: Department of Physics and Astronomy, University of British Columbia, Vancouver, BC, V6T 1Z1, Canada.

[1] T. Ditmire *et al.*, Nature (London) **386**, 54 (1997).
[2] M. Lezius *et al.*, Phys. Rev. Lett. **80**, 261 (1998).
[3] V. Kumarappan, M. Krishnamurthy, and D. Mathur, Phys. Rev. Lett. **87**, 085005 (2001).
[4] T. Ditmire *et al.*, Nature (London) **398**, 489 (1999).
[5] J. Zweiback *et al.*, Phys. Plasmas **9**, 3108 (2002).
[6] G. Grillon *et al.*, Phys. Rev. Lett. **89**, 065005 (2002).
[7] K. W. Madison *et al.*, Phys. Plasmas **11**, 270 (2004).
[8] T. Ditmire *et al.*, Phys. Rev. A **53**, 3379 (1996).
[9] U. Saalmann and J.-M. Rost, Phys. Rev. Lett. **91**, 223401 (2003).
[10] S. Sakabe *et al.*, Phys. Rev. A **69**, 023203 (2004).
[11] I. Last and J. Jortner, Phys. Rev. Lett. **87**, 033401 (2001).
[12] I. Last and J. Jortner, J. Phys. Chem. A **106**, 10 877 (2002).
[13] I. Last and J. Jortner, J. Chem. Phys. **121**, 8329 (2004).
[14] K. Ishikawa and T. Blenski, Phys. Rev. A **62**, 063204 (2000).
[15] C. Jungreuthmayer, M. Geissler, J. Zanghellini, and T. Brabec, Phys. Rev. Lett. **92**, 133401 (2004).
[16] B. N. Breizman, A. V. Arefiev, and M. V. Fomyts'kyi, Phys. Plasmas **12**, 056706 (2005).
[17] J. Purnell *et al.*, Chem. Phys. Lett. **229**, 333 (1994).
[18] E. M. Snyder *et al.*, Chem. Phys. Lett. **248**, 1 (1996).
[19] J. W. G. Tisch *et al.*, Phys. Rev. A **60**, 3076 (1999).
[20] O. F. Hagena and W. Obert, J. Chem. Phys. **56**, 1793 (1972).
[21] R. A. Smith, T. Ditmire, and J. W. G. Tisch, Rev. Sci. Instrum. **69**, 3798 (1998).