Anisotropic Explosions of Hydrogen Clusters under Intense Femtosecond Laser Irradiation

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We report on measurements of ion energy distributions from hydrogen clusters irradiated by intense laser pulses of duration 40 and 250 fs. Contrary to the predictions of a simple Coulomb explosion model, we observe a pronounced spatial anisotropy of the ion energies from these explosions with the highest energy ions ejected along the laser polarization direction. The origin of the anisotropy is distinct from that previously seen in clusters of high Z atoms such as Ar and Xe. Furthermore, a measured increase in H^+ ion energy when longer, lower intensity pulses are employed suggests that multiple-pass, vacuum heating of the cluster electrons is important in the deposition of energy by the laser.

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Gases of atomic and molecular clusters are of continuing interest as targets for intense laser-matter interactions because of their unique absorption and optical properties [1–4]. Clusters subjected to strong laser fields can efficiently absorb laser energy leading to the ejection of fast ions [5–11], an effect which has been exploited to produce nuclear fusion in gases of deuterated clusters [12,13]. The heating of the cluster by the intense laser and the subsequent explosion can often be described by one of two simple models. If the laser fully strips the cluster of its electrons, the ions explode isotropically through a Coulomb explosion. The cluster is said to undergo cluster vertical ionization (CVI) if complete stripping of the cluster electrons occurs before any significant ionic motion [14]. On the other hand, in large clusters, if the majority of the free electrons are retained by the space-charge field of the cluster ions, the cluster can be pictured as a quasineutral plasma sphere which undergoes an isotropic hydrodynamic expansion [1,2]. In many situations, however, the cluster is partially stripped of its electrons and neither of these limiting cases will appropriately describe the explosion dynamics. In this case, the electrons can develop two distinct populations [15,16]: a bound electron core and a cloud of extracted electrons, both of which move with a preferential direction along the laser field. Unlike the Coulomb explosion and hydrodynamic model, the spherical symmetry of the cluster expansion is broken in this intermediate regime.

Anisotropy has been observed by a number of groups in ion energy spectra measurements from large (>1000 atoms/cluster) N_2, Ar, and Xe clusters irradiated at modest intensity ($\sim 10^{16}$ W/cm^2) [7–9,17,18]. There has been substantial debate regarding the mechanisms for this anisotropy. It has been surmised that the laser induced surface polarization on high Z clusters could lead to preferential acceleration of surface ions along the laser field in the hydrodynamic limit [17]. Recent results on anisotropic explosions in Ar clusters by Kumarappan *et al.* [7] were explained as a consequence of two effects: the production of higher charge state ions near the poles of the cluster resulting from enhanced field ionization there and direct laser field acceleration of these highly charged ions through a process termed "charge flipping" [8]. This interpretation was strengthened by microscopic particle-in-cell simulations of the explosion of large ($N > 10\,000$) clusters of multielectron atoms by Jungreuthmayer *et al.* [19]. In this model, as the electron cloud is pulled out of the cluster by the laser field, the opposite pole is depleted. This leads to an enhanced electric field which can yield higher charge states at the poles of the cluster. These more highly charged ions can attain superior kinetic energies in the explosion leading to a hardening in the ion spectrum in the laser field direction. In addition, the charge state of the ions at the poles vary every laser half-cycle through a cyclical process of laser field ionization and rapid recombination, leading to a cycle averaged net force along the laser field axis.

Both of these physical mechanism *cannot* play a role in the explosions of hydrogen clusters at high intensity because all atoms are completely ionized very early during the laser irradiation. Previous studies of hydrogen cluster explosions under intense laser irradiation have led to ion spectra which are consistent with the CVI model followed by Coulomb explosion. For example, Sakabe *et al.* [10] irradiated small H_2 clusters (≈25 Å) at 6×10^{16} W/cm^2 finding that the energy spectra agree well with the fully stripped Coulomb explosion model. Our group has previously examined deuteron energy spectra from a dense D_2 cluster medium irradiated with 100 fs pulses of peak intensity $\sim 2 \times 10^{20}$ W/cm^2 also finding that energy spectra were consistent with a pure Coulomb explosion prediction [20]. Both of these studies, however, did not consider the angular distribution of the ejected ions from the clusters or the details of the shape of the ion spectra at the highest energies, both quantities of which are sensitive to deviations from the simple Coulomb explosion description of the explosions.

In this Letter, we measure the angular dependence and detailed energy spectra of ions ejected from large H_2 clusters irradiated at high intensity. We see a clear spatial asymmetry in these explosions with ion energies enhanced along the laser polarization direction. This effect cannot be explained by the model invoked in references [7,19] which is predicated on spatially varying ionization state of ions in the cluster. Instead, we believe that in these $Z = 1$ clusters, the oscillating electron cloud during cluster ionization gives rise to the observed ion anisotropy, as predicted by the theory of Breizman *et al.* [16]. We conducted measurements for both short (40 fs) and long (250 fs) pulses focused to nearly 10^{18} W/cm^2. We find that the measured ion energies and spectral shapes near the peak ion energy can be reasonably well explained using a CVI Coulomb explosion model while the lower energy ions, which presumably arise from deeper in the cluster, deviate from this model, consistent with the predictions of a picture based on partial stripping of the cluster and the formation of an oscillating inner cold electron core. We find, in addition, that higher ion energies were attained with the longer pulse, lower intensity irradiation which we interpret as evidence for the vacuum heating of cluster electrons and more complete evacuation of the cluster [4,15,16,21].

To conduct this experiment we irradiated a low density stream of clusters formed from a cryogenically cooled pulsed supersonic gas jet [22] with an orifice-diameter of 750 μm directed through a skimmer. The energies of the ions created by the laser cluster interaction were determined by measuring the time of flight (TOF) of the ions in a field free drift tube of 1.14 m length. A microchannel plate detected the ions ejected perpendicular to the laser propagation in an angular cone of 7.8×10^{-4} sr. We estimated the average cluster radius as $\langle R_0 \rangle \approx 20$ Å using the Hagena scaling law [23] $\Gamma^* = k p_0 (D/\tan\alpha)^{0.85}/T_0^{2.29}$ where α is the expansion half angle (5°), $T_0 = 100$ K and $p_0 = 250$ psi are the reservoir temperature and pressure, and $k = 184$ for H_2 bonding. We irradiated the clusters with the 800 nm THOR Ti:sapphire laser system operating in two configurations: a pulse energy of $\sim$30 mJ and full width half maximum (FWHM) duration 40 fs, and a pulse energy of $\sim$100 mJ and duration 250 fs (created by chirping the shorter pulse), both focussed to a peak intensity of ≈ 0.5–1.0×10^{18} W/cm^2. The laser polarization was linear and rotated using a half-wave plate before the focusing lens.

Measured ion energy spectra are shown in Fig. 1 for two orthogonal polarizations, 0° (black) and 90° (gray), where 0° is defined as the laser electric field pointing toward the detector. There is a clear hardening of the ion spectra for both pulse durations over the ion energy range of 1 to 8 keV for ions ejected along the laser polarization (0°). With a 40 fs pulse we observe $E_{av} \approx 1$ keV at 0° and $E_{av} \approx 0.6$ keV at 90° while with 250 fs pulses we observe higher ion energies with $E_{av} \approx 2$ keV at 0° and $E_{av} \approx 1.6$ keV at 90°. We do not, however, observe a sizable difference for either pulse duration in the *maximum* ion energy for the two polarizations. The most significant aspect of the data in Fig. 1 is the sizable difference in the

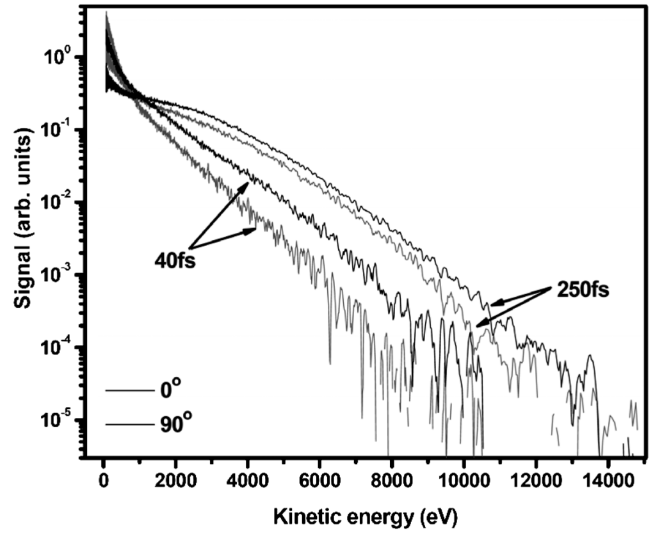

FIG. 1. Proton kinetic energy spectra from H_2 clusters irradiated with 40 and 250 fs laser pulses with 0° (black) and 90° (gray) polarization.

average observed energy between ions ejected along the laser polarization and those ejected perpendicular to the polarization. The mean energies as a function of polarization angle for both pulse durations are shown in Fig. 2. In both cases the ion energy is maximum at 0° with the degree of anisotropy, $[E_{av}(0°) - E_{av}(90°)]/E_{av}(0°)$, being $\approx$40% with 40 fs and $\approx$25% with 250 fs. Also striking in these data is the large increase in energy resulting from the use of the longer 250 fs pulse.

These data are at odds with the Coulomb explosion picture usually thought to be appropriate for hydrogen clusters. This picture, which is most appropriate for low

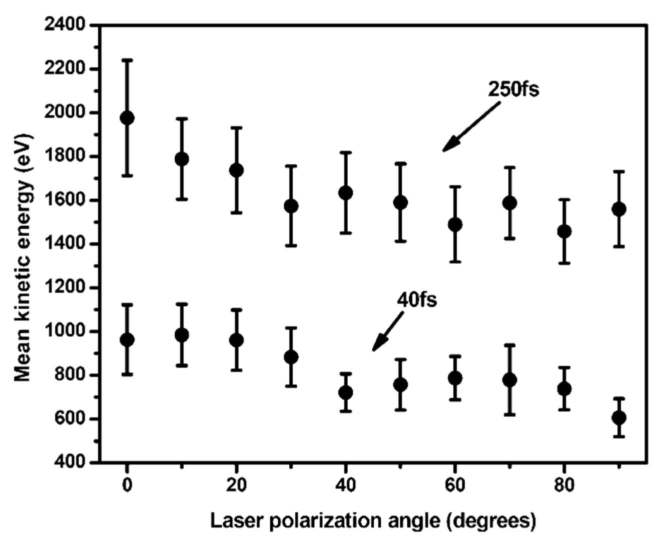

FIG. 2. Average proton energies from H_2 clusters irradiated with 40 and 250 fs laser pulses as a function of observation angle where 0° represents observation along the direction of the laser electric field.

Z clusters subject to high peak ponderomotive potential, U_p, well above the peak electrostatic field confining the electrons, ϕ_{confine}, predicts an isotropic explosion. Furthermore, the increase in ion energy from longer pulse irradiation is contrary to what would be expected when longer pulses are employed in the Coulomb explosion regime. We have, for example, previously observed indirect evidence for a decrease in the energy of ions in deuterium clusters irradiated with 100 fs to 1 ps pulses. Those experiments were conducted at very high intensity, $>10^{20}$ W/cm^2, so the clusters were expected to be completely stripped of all their electrons very early in the pulse [24]. Similar conclusions were found in the calculations of Ref. [14].

The explanation for anisotropy of Ar clusters described in Refs. [7,8] rely on a spatially varying ionization state in each cluster cannot be at work in our experiment of $Z = 1$ hydrogen ions. In fact, both observations, the enhancement of ion energies along the laser polarization and the increase in ion energy with longer pulses, can be explained by the formalism of Breizman *et al.* [16] which treats the case in which the cluster is only partially stripped of electrons. In this model a cold electron core of unextracted electrons develops in the cluster sphere when the laser field is comparable to or less than the electrostatic confining field of the cluster ions, ie when $U_p \sim \phi_{\mathrm{confine}}$ (This situation is roughly true in our experiment; while the peak U_p is $\sim$50 keV, it will be considerably less over the majority of the focal region and during the rising part of the pulse. This U_p is comparable to ϕ_{confine} for the largest clusters in our distribution where $\phi_{\mathrm{confine}} \sim 30$ keV). The electrons remaining inside the cluster form a cold spherical electron core smaller than the ion radius that remains quasineutral within the positively charged outer ion shell. In the early stages of the interaction when the cluster plasma density is high, the plasma frequency is much higher than the laser frequency; the core of electrons responds adiabatically to the laser field, remains in equilibrium, and oscillates at the laser frequency. As the laser intensity increases, the laser field removes electrons from the cluster until the core displacement reaches a maximum value. If the cluster radius is much smaller than the excursion distance of an electron in the laser field (also true in our experiment), the extracted electrons escape from the cluster vicinity leaving a neutral core surrounded by a positive ion shell. In this regime the ion acceleration of the outer non-neutral ion shell is driven solely by the Coulomb forces. The spherical symmetry of the explosion is broken by the fact that the inner electron core oscillates within the ion sphere. It was shown in Ref. [16] that if a sizeable fraction of electrons is extracted (in other words, that the cold electron core is significantly smaller than the ion sphere radius) the ions at the surface of the cluster, those that yield the highest energy upon explosion, feel a time averaged radial accelerating potential that is nearly isotropic while those ions deeper in the cluster feel an accelerating field that is higher along the axis of the laser polarization.

This prediction is completely consistent with our data. We find, as illustrated in Fig. 1, that for both pulse durations there is very little difference in the cutoff energy of the highest energy ions, those ions presumably ejected from near the surface of the largest clusters, for both polarization directions. However, there is significant shift of ions from low energy ($<$1 keV) to intermediate energies (2–8 keV) in the spectra when the observation direction is shifted from perpendicular to parallel with the laser E field. These lower energy ions probably come from deeper in the clusters where the cold electron core theory predicts that the time averaged accelerating field is higher along the laser field.

This conclusion, which relies on the presence of a cold electron core within a Coulomb exploding outer ion shell is further reinforced when we compare our measured spectra to the predictions of a simple, pure Coulomb explosion of fully stripped clusters. In this regime, the energy distribution of ions from a CVI-initiated Coulomb explosion of a single cluster size is simply $f(E)dE \propto \sqrt{E}dE$ for energies less than the maximum energy acquired by ions at the surface, i.e., $E \leq E_M = n_H e^2 R_0^2/3\varepsilon_0$, where n_H is the initial ion density ($\approx 5 \times 10^{22}$ atoms cm^{-3}) and R_0 is the cluster radius. To obtain an estimate for the observed ion spectrum, we convolve this single cluster spectrum with the cluster size distribution [20]. We used our expected $\langle R_0 \rangle \approx 20$ Å to construct a log-normal distribution of cluster sizes of the form $f(N) \propto \exp[-\ln^2(N/N_0)/2w^2]$ where N_0 is the modal cluster size (number of atoms) and w is proportional to the FWHM of the distribution. The energy distribution function resulting from irradiation of this ensemble of clusters is $F(E)dE \propto g(E)\sqrt{E}dE$, where $g(E) = \int_{N_\varepsilon}^{\infty} n_C(N)dN$ is the density of clusters larger than the size N_E for which $E_M = E$. This approach has been successful in reproducing deuterium cluster ion spectra from very high intensity 100 fs irradiation [20].

The energy spectrum generated with $w = 0.88$ is compared to our data in Fig. 3. The cluster size distributions with the same $\langle R_0 \rangle$ generated with $w = 0.25$ and $w = 0.5$ are also shown in the inset. The shape of the CVI spectrum with $w = 0.88$ matches well the hot ion tail of the 250 fs pulse generated distribution at ion energies between 8 and 13 keV. These high energy ions arise from the near the surface of the cluster ion shell and should approximate the pure Coulomb explosion spectrum if the cold ion core is smaller than the cluster radius, when the cluster is significantly, if not totally, stripped of its electrons (a situation expected in our experiment at the highest intensity, where $U_p \sim \phi_{\mathrm{confine}}$). There is, however, a deviation from the pure CVI spectrum in the data at the intermediate ion energy range of 2 to 8 keV. In this region, there is a abundance of ions compared to the calculated distribution while below this energy (i.e., 0–2 keV) there is a dearth of ions. This behavior is particularly evident in the longer pulse data where these interior ions feel the effects of the time averaged field from the oscillating electrons for more

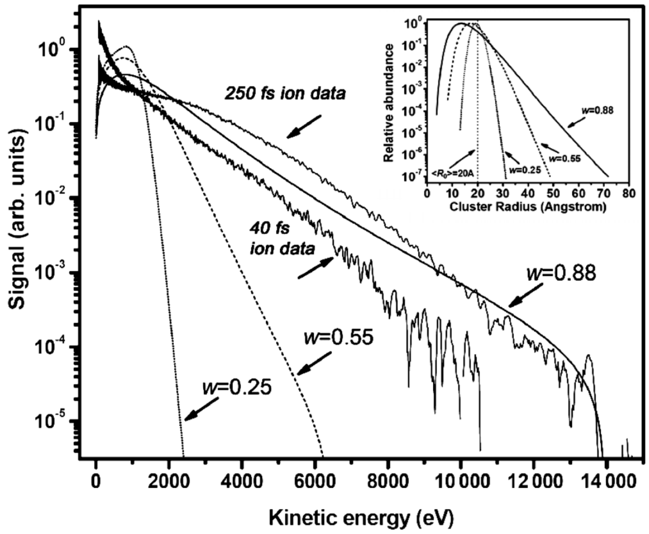

FIG. 3. Energy distributions of protons calculated using the vertical ionization Coulomb explosion model assuming log-normal cluster size distributions (shown in the inset), compared with the 0° polarization experimental data of the two pulse lengths 40 and 250 fs.

laser cycles. This seems to be consistent with the fact that the larger clusters in our experimental distribution, where 30 Å $< R_0 <$ 60 Å, those that contribute predominately to the ions with energy $>$2 keV, the clusters are only partially stripped. Only a small fraction of electrons are retained within the cluster at our peak intensity ($<$10% for $R_0 =$ 60 Å). This small core of unextracted electrons will not greatly affect the high energy ions which are accelerated from the outer layers of the cluster but will affect the inner ions.

Finally, the clear shift of the measured ion spectrum to higher energies for both polarizations when longer pulses are employed is a consequence of the vacuum heating of the cold inner core of electrons. As described in Ref. [16], the cold electron core, which cannot be directly extracted by the laser, can experience stochastic heating as these electrons oscillate near the ion sphere surface. As they acquire energy in this manner they can ultimately escape the cluster. When longer pulses are used, the greater number of laser cycles drives superior vacuum heating and more efficient extraction of the inner core of electrons. This effect is significant even though the clusters are expanding on the time scale of the longer pulse. The characteristic time scale for the expansion of a fully stripped hydrogen cluster is roughly an inverse plasma ion period; i.e., $t_{\text{expand}} \approx (3m_{\text{ion}}\varepsilon_0/e^2 n_{\text{cluster}})^{1/2}$, which is $\sim$10 fs for hydrogen clusters [24]. Even though the largest clusters are not completely stripped of electrons, we do expect significant expansion for both pulse durations, particularly for the 250 fs pulses. That we observe higher ion energies when the longer pulses are employed indicates that stochastic heating is important in removing additional electrons from the largest clusters in the distribution.

In conclusion, we have measured ion energy distributions from explosions of H_2 clusters irradiated at intensity up to 10^{18} W/cm^2. We find angular distributions of ions that are enhanced along the laser polarization, similar to anisotropy observed previously from Ar cluster. Unlike these earlier results which were ascribed to an anisotropy in the ionization state of the ions with the cluster, we interpret our data in light of incomplete stripping of the clusters by the field and the result of the oscillating electron cloud around the cluster. Furthermore, we find that longer pulses actually increase the average ion energies of the exploding clusters, a consequence of the greater vacuum heating that electrons can undergo in the longer pulse.

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