Measurements of ion energies from the explosion of large hydrogen iodide clusters irradiated by intense femtosecond laser pulses

J. W. G. Tisch, N. Hay, E. Springate, E. T. Gumbrell, M. H. R. Hutchinson, and J. P. Marangos
Blackett Laboratory Laser Consortium, Imperial College of Science, Technology and Medicine, London SW7 2BZ, United Kingdom
(Received 8 June 1999)

We present measurements of ion energies from the interaction of intense, femtosecond laser pulses with large mixed-species clusters. Multi-keV protons and ~100-keV iodine ions are observed from the explosion of HI clusters produced in a gas jet operated at room temperature. Clusters formed from molecular gases such as HI are thus seen to extend the advantages of the laser-cluster interaction to elements that do not readily form single-species clusters. In the light of recently reported nuclear fusion in laser-heated clusters, we also examine the possibility of boosting the explosion energies of low-Z ions through the use of mixed species clusters. [S1050-2947(99)03310-7]

PACS number(s): 36.40.Gk, 52.50.Jm

Over the last few years, experiments conducted by a number of groups have shown that the interaction of short (<1 ps), intense ($\sim 10^{16}$ W cm^{-2}) laser pulses with clusters of more than a few hundred atoms produced in gas jets can result in debris-free, bright keV x-ray generation [1,2], and the production of extremely energetic electrons [3] and highly charged fast ions [4,5]. Many aspects of the laser-cluster interaction have been successfully described by a numerical plasma model [6], which treats the ionized cluster as a uniform, spherical nanoplasma.

The nanoplasma model shows that the interaction can be divided essentially into three phases. The first is the initial field ionization of the near-solid-density, subwavelength scale cluster that occurs early on the rising edge of the high-intensity laser pulse. Owing to the high density of the nanoplasma, once some electrons have been produced the second phase is dominated by collisional ionization and collisional heating. The ions are stripped to high charge states and the electrons are efficiently heated by the laser, in the absence of conduction to any bulk material.

The third phase is the ''explosion'' of the cluster. This is triggered when the electron density in the expanding cluster plasma has dropped to three times the critical density, leading to a dielectric enhancement of the laser field inside the nanoplasma relative to the external field. This resonance produces a sharp increase in the electron heating that leads to a very rapid expansion (explosion) of the cluster plasma, driven largely by the hydrodynamic pressure of the hot electrons, according to our simulations and experimental results. In the hydrodynamic expansion, the initially cold, highly stripped ions are rapidly accelerated to high kinetic energies in the ambipolar potential set up by the expanding electrons. Our modeling shows that the ions ultimately attain an average energy in the region of $\langle Z_i \rangle k_b T_e$, where $\langle Z_i \rangle$ is the average ion charge state, k_b is Boltzmann's constant, and T_e is the electron temperature at the resonance. Experimentally, for high Z clusters, the ion energy distribution has also been found to exhibit a much higher-energy tail, containing a small number of ions with energies much greater than $\langle Z_i \rangle k_b T_e$, attributed to the Coulomb ejection of highly charged ions near the surface of the cluster [5].

In the context of heating by high-intensity lasers, mixed species clusters have to date received little attention, but are interesting for a number of reasons. First, clusters from molecular gases potentially extend the advantages (i.e., extremely efficient, debris-free transfer of laser energy to particles and x-ray photons) of the laser-cluster interaction to elements that are hard to cluster in the pure form, such as atoms and molecules with a low propensity for cluster formation. For example, large-cluster formation in gas jets of hydrogen (and its isotopes) requires very high pre-expansion pressures and cryogenic cooling because the condensation parameter for hydrogen is very low [12]. Hydrogen iodide gas, on the other hand, forms large clusters very readily at low pressures without any cooling.

Second, a high $\langle Z_i \rangle$ is required for efficient collisional heating of the nanoplasma electrons. According to our model, the maximum ion energy from the explosion of single-species clusters of the same number of atoms scales as $\sim \langle Z_i \rangle^2$ (see Fig. 1). Therefore, in a cluster comprising low and high Z species, the more highly stripped high Z ion species should allow a higher T_e to be attained to drive the explosion of both ion species. Further, in the expansion of a two-ion species plasma for the planar case, it is known that under certain circumstances (depending on the relative ion masses, charges, and densities), one species can attain a

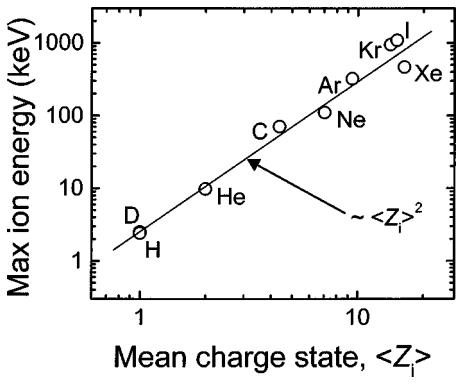

FIG. 1. Simulated scaling of the maximum ion energy with the average charge state of the cluster plasma, $\langle Z_i \rangle$, for 60 000-atom clusters irradiated by 260-fs, 780-nm pulses at 2×10^{16} W cm^{-2}.

much higher energy than the other, with a small fraction of these ions achieving energies of up to $\sim 1000T_e$ [7]. Given the success of the plasma model for the laser-heated cluster, it is not unreasonable to expect a similar behavior in a mixed-species cluster. Boosting the explosion energies of low Z ions in a two-species cluster would be highly relevant to cluster-fusion research, in the light of the recent demonstration of nuclear fusion in laser-heated deuterium clusters [8].

This is not the first investigation of HI clusters in a strong laser field. Purnell *et al.* [9] reported ion kinetic energies up to 700 eV and charge states up to I^{8+} in the interaction of 150-fs, 624-nm laser pulses at 10^{15} W cm^{-2} with small HI clusters (<10 atoms). These energies were explained in terms of a pure Coulomb explosion of the multiply charged HI clusters. No measurements of the proton energies were reported. In our paper, much larger HI clusters, for which the nanoplasma model is applicable, were irradiated at higher intensity and time-of-flight (TOF) measurements were made of the proton and I^{n+} kinetic-energy distributions.

The laser system [10] and TOF spectrometer [11] used for this paper have been detailed elsewhere. The HI clusters were produced in a gas jet from a pulsed valve [12] backed with HI gas (3–7.8 bar). A skimmer was used to produce a cluster beam with a density $<10^{11}$ clusters/cm^3 in the interaction region. To estimate the size of the HI clusters, we use Hagena's reduced-scaling parameter Γ^* [13], which correlates cluster formation in jets of different gases. Gas expansions with the same Γ^* tend to cluster to a similar degree. Using Eq. 12 in Ref. [13], we estimate the ratio $\Gamma^*(\mathrm{HI})/\Gamma^*(\mathrm{Xe}) \approx 2.2$, and since the number of particles per clusters, N_c, scales like $(\Gamma^*)^2$, we expect $N_c(\mathrm{HI}) \approx 5N_c(\mathrm{Xe})$ for the same gas-backing pressure. From our previous calibration of $N_c(\mathrm{Xe})$ as a function of backing pressure for this pulsed valve (using Rayleigh scattering measurements), we infer $N_c(\mathrm{HI}) \approx 60\,000$ molecules at 7.8-bar-backing pressure.

The clusters were irradiated by 260-fs pulses from a 10-Hz Ti:sapphire (780 nm) chirped pulse amplification laser, focused at $f/8$ to an intensity above 10^{16} W cm^{-2}. A background pressure of 10^{-7} mbar was maintained in the interaction chamber. The ions produced in the cluster explosions were detected by a microchannel plate (MCP) detector (Galileo APTOF-18), positioned 58 cm from the interaction region. The MCP signal was acquired by a digital oscilloscope and the TOF spectrum for each shot was downloaded to a PC in real time for processing.

No extraction voltage was employed to accelerate the ions towards the MCP, so the time of flight to the detector provided a direct measurement of the kinetic energy of the ions from the explosion of isolated HI clusters. However, before impacting the detector, the ions were accelerated in a 2-mm region between a grounded grid at the end of the flight tube and the input face of the MCP, which was at a potential of $V=-2$ kV. This has a negligible effect on the flight time from which the ion kinetic energies are calculated, but modifies the ion impact energy distribution according to $f(E) \rightarrow f(E+VZ_i)$. This serves to compress the impact energy range, e.g., for protons with initial energies in the range 0.1–2.5 keV, from a factor of 25 to a factor of 2, which makes the MCP response more uniform. The ion charge states were not resolved in this experiment.

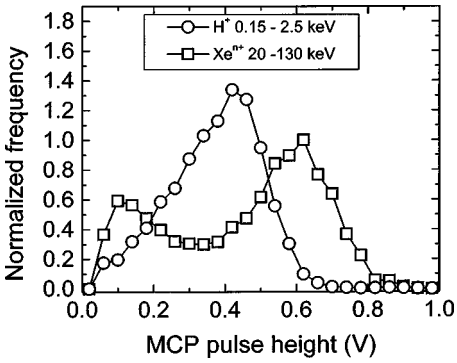

FIG. 2. Averaged MCP pulse-height distributions for protons with energies in the range 0.15–2.5 keV (round points) and multiply charged Xe ions with the same range of flight times (squares).

An aspect of this experiment was the use of a pulse-height analysis of the MCP signal to differentiate between hits from protons and I^{n+} [necessary because I^{n+} and H^+ ions with energies in the ratio of their masses (127:1) have the same TOF]. This was based on separate experiments we conducted that showed that protons with initial energies in the range 0.15–2.5 keV (covering the range of proton energies from the HI clusters) exhibit a quite different pulse-height distribution to fast Xe ions with a similar range of flight times (i.e., $\sim 130\times$ more energetic), as shown in Fig. 2. The proton pulse-height distributions were obtained by using an extraction voltage in the range 0.3–5 kV to accelerate initially cold (<50 eV) protons out of the interaction region following laser ionization (just above the appearance intensity) and dissociation of contaminant H_2O in the interaction chamber. Note that without an extraction voltage, these background protons do not reach the MCP. The Xe pulse-height distributions were acquired using energetic Xe ions (20–200 keV) from the explosion of Xe clusters. Xe was used owing to the technical difficulties involved in producing pure I clusters. The pulse-height response of MCPs is generally not very well understood, so the origin of the difference in the Xe^{n+} and proton pulse-height distributions is not known at this stage. It may be due to the much higher average charge state of the Xe ions—known to be in the range $10+-30+$ [11]—or their higher energies.

The application of this pulse-height discrimination technique to distinguish I^{n+} from protons is validated by our observation of the characteristic pulse-height distributions described above (Fig. 2) in the ion TOF spectra from HI cluster explosions. Figure 3(a) shows the raw pulse-height resolved TOF spectrum for a HI backing pressure of 7.8 bar and a laser intensity of 2×10^{16} W cm^{-2}. The spectrum was accumulated from ~ 2000 consecutive laser shots, with approximately 10 ion hits per shot. Three features can be seen. The partially resolved feature at early time (0–0.5 μs) labeled (i) is due to very fast particles, which we expect to be mostly electrons [3]. Feature (ii) is the grouping of hits arriving between $\sim 1-3.5$ μs that has a pulse-height distribution that is clearly different from that of (iii) that comprises hits arriving largely after ~ 3 μs, though there is some temporal overlap between these two. Importantly, (ii) and (iii)

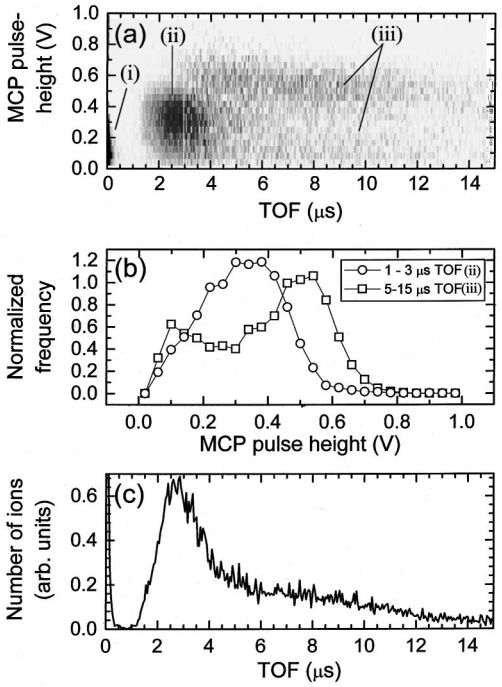

FIG. 3. (a) Raw pulse-height-resolved TOF spectrum for HI cluster explosions at $2\times10^{16}\,\mathrm{W\,cm^{-2}}$ and 7.8-bar pulsed-valve backing. (i), (ii), and (iii) label three distinct features, attributed, respectively, to fast electrons, protons, and multiply charged iodine ions. (b) Averaged pulse-height distributions of features (ii) (circles) and (iii) (squares). (c) TOF spectrum corresponding to data in (a) integrated over all pulse heights.

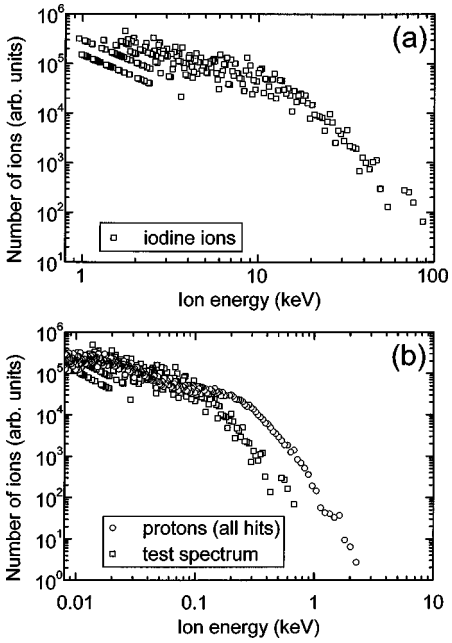

FIG. 4. (a) I^{n+} kinetic-energy spectrum from the explosion of 60 000-molecule HI clusters at $2\times10^{16}\,\mathrm{W\,cm^{-2}}$. (b) Corresponding proton spectrum calculated from all measured ion hits (circles). The low-energy portion of this spectrum (<100 eV) is due to the erroneous inclusion of I^{n+} hits, as seen in the comparison with a vertically scaled test spectrum (squares) of known iodine ions treated as protons.

have pulse-height distributions—displayed in Fig. 3(b)—that are only weakly dependent on the TOF. The close similarity of these with the proton and Xe ion pulse height distributions (Fig. 1) leads us to the conclusion that feature (ii) is due to protons, and feature (iii) is due to multiply charged iodine ions. The resemblance between the I^{n+} and Xe^{n+} pulse-height distributions is not surprising given the near-identical ion masses and the expected similarity of their charge state and energy distributions. Figure 3(c) shows the same TOF spectrum integrated over all pulse heights. Without the pulse-height information, the only hint to the presence of the two ion features is the shoulder in the spectrum not normally seen in the typical spectrum from a pure rare-gas cluster explosion [11].

In Fig. 4 we display the ion kinetic-energy spectra corresponding to the data in Fig. 3(a). Taking I^{n+} hits to be those with a pulse height greater than 0.6 V enables us to extract the I^{n+} energy spectrum, shown in Fig. 4(a). Since the I^{n+} pulse-height distribution is only a weak function of the TOF, the energy spectrum calculated from this selection of hits should closely approximate the true spectrum. The maximum kinetic energy is ~92 keV, while the mean of the distribution is ~8.6 keV. The round points in Fig. 4(b) show the proton-energy spectrum obtained by assuming that all measured ion hits are due to protons. Clearly, half these hits are actually from I^{n+}. We show that the iodine ions introduce an error only to the low-energy part of the spectrum (<100 eV) by plotting the energy spectrum of the known I^{n+} hits (pulse height >0.6 V) treated as protons, i.e., using the proton mass in the calculation of the kinetic energy. This test spectrum (square points) has been scaled vertically to reflect the fact that the I^{n+} hits comprise half of all the measured ion hits. Comparing the round and square points, it is apparent that spectrum below 100 eV is due almost exclusively to the erroneous inclusion of I^{n+} hits, while the spectrum above 100 eV is due to protons. Thus, taking 100 eV as the lower limit of the proton spectrum, the mean and maximum proton energies are approximately 270 eV and 2.5 keV, respectively. We observed a monotonic increase in both I^{n+} and proton energies with increasing HI cluster size up to a maximum dictated by the maximum pressure available from our regulator (7.8 bar).

The observation of energetic protons and iodine ions from the laser heating of HI clusters, while perhaps not surprising, proves that molecular clusters can be used to produce energetic ions from a range of elements much broader than those that cluster readily in the pure form. For example, highly charged, heavy ions ($A\approx200$) are required for seeding particle accelerators. Traditional production methods rely on solid-target interactions using large-scale, long-pulse CO_2 lasers [14]. A compact source of highly stripped tungsten ions, for example, might be possible from the explosions of clusters of WF_6 (a gas at room temperature) that could be formed in a standard gas jet—a much easier route than trying to form the clusters from the refractory metal.

Theoretical analysis of the energetics of the laser-HI cluster interaction is difficult for two reasons. First, because a mixed-species laser-cluster model does not exist at the present time (and there is no self-consistent way of extending our nanoplasma model to include more than one ion species). Second, because the direct comparison with the ion energies from pure H and I clusters was not attempted in this experi-

ment owing to the extreme technical difficulties involved. For example, to make 60 000-atom H_2 clusters would require cooling to well below liquid-nitrogen temperatures [12], which emphasizes the advantages of using HI gas for the formation of clusters containing hydrogen.

However, to make a comparison with the ion energies from a pure cluster, we measured the ion spectrum from the explosion of 5000-atom Xe clusters under identical experimental conditions. A maximum Xe ion energy of ~150 keV was recorded, which, from our simulations, we take to be a lower limit for the I^{n+} energy from the explosion of a 5000 atom pure I cluster. This result indicates that the maximum ion energy from a pure 60 000-atom I cluster would be significantly greater than the ~90 keV measured from the 60 000-molecule HI cluster (based on the monotonic increase in the ion energy with increasing cluster size between 5000 and 60 000 atoms that is predicted by our model).

The lower iodine ion energies from the mixed species cluster can be attributed, at least in part, to the lower effective $\langle Z_i \rangle$ in the HI nanoplasma owing to half the ions being only singly charged. This will reduce the electron heating which in turn will modify the thermal collisional ionization and hence $\langle Z_i \rangle$ for the iodine ions. The strong interdependence of these variables makes this effect difficult to quantify in the absence of a two ion species nanoplasma model. The plasma conditions at the onset of the explosion also depend on the initial density of the neutral cluster, since the initial density affects the timing of the resonance relative to the peak of the laser pulse. Our modeling shows that T_e and $\langle Z_i \rangle$ are actually very similar for 60 000-atom I clusters at I or HI densities for a laser pulse duration of 260 fs, but this is not true in the general case. Thermalization between the electrons and the protons during the cluster-heating phase would also reduce the electron temperature in the HI cluster. However, for $k_b T_e$ greater than a few tens of eV, the electron-proton equilibration time is significantly longer than the ~100-fs time scale of the collisional heating, so there is negligible transfer of energy from the electrons to the protons during the heating phase.

The final ion energies also depend on the dynamics of the plasma expansion. The self-similar expansion of a collisionless two-ion species plasma has been analyzed in the literature [7,15]. It is known that the ion species can separate in the expansion, owing to the difference in the charge-to-mass ratios. Such a separation is evident in our TOF data shown in Fig. 3(a), with the majority of the protons reaching the detector before the iodine ions. Gurevich, Pariiskaya, and Pitaevskii [7] also showed that a small fraction of impurity ions can acquire energies of up to $10^2 - 10^3\, k_b T_e$ in one-dimensional, two-species expansions. It is assumed that the impurity ions do not affect the motion of the other ion species (they are not included in the ambipolar potential). The dynamics of a plasma expansion into vacuum is known to depend strongly on the geometry of the system [16]. However, if we apply this planar analysis to a spherical cluster plasma we find that the protons in the HI cluster explosion can initially be treated as impurity ions, despite equal ion densities, because the impurity condition, $Z_2 N_2 \ll Z_1 N_1$ (where 1 and 2 refer to I^{n+} and H^+, respectively, and N_i is the density), is satisfied since $Z_1 \gg Z_2$ (for a pure iodine cluster of 60 000 atoms and the same laser conditions, $Z_1 \approx 15$, according to our simulations).

Treating the protons as impurity ions has two important consequences. First, the motion of the iodine ions is not modified in the expansion compared to the single-species case. The difference in the maximum I^{n+} energy from the mixed and single-species cluster is thus a consequence of the different initial conditions of the explosion. Second, protons with energies of the order of 100 keV might be expected for $k_b T_e > 1$ keV. In fact, we cannot rule out the existence of very energetic protons on the basis of our data, since they would be masked in our TOF spectra by the electron peak [feature (i) in Fig. 3(a)]. We note that for fusion in deuterium iodide clusters, a small fraction of very fast deuterons might have a significant effect on the fusion yield, given the very strong scaling of the fusion cross section with deuteron energy. Clearly, further work is necessary to establish if protons of such energy are being produced.

In conclusion, we have measured the I^{n+} and proton kinetic energies from the explosion of HI clusters (estimated size, 60 000 molecules) irradiated at $2 \times 10^{16}\,\mathrm{W\,cm^{-2}}$ by 780-nm, 260-fs pulses. We find that the I^{n+} energies are much higher than those observed previously in laser interaction with smaller HI clusters, but significantly less than those expected from the explosion of a pure I cluster with the same number of atoms. Qualitatively, the reduction is consistent with a reduced average ion charge state in the HI nanoplasma. It is not clear yet whether the H^+ energies are boosted compared to a pure H cluster explosion in the two-ion-species expansion, but theoretical work on two-ion-species plasma expansions suggests that such an enhancement should be possible—with very high proton energies predicted. The use of molecular clusters also permits the efficient laser heating of many elements that do not readily form clusters in the pure form. Certainly, the laser heating of mixed species clusters is a topic deserving further experimental and theoretical investigation.

The authors are grateful to R. A. Smith and J. P. Connerade for useful discussions. Help with the laser and experiment from M. B. Mason and K. J. Mendham and the technical assistance of P. Ruthven and A. Gregory are also gratefully acknowledged. This work is funded by the EPSRC and J.W.G.T. is supported by the EPSRC.

[1] A. McPherson *et al.*, Nature (London) **370**, 631 (1994).
[2] S. Dobosz *et al.*, Phys. Rev. A **56**, R2526 (1997).
[3] Y. L. Shao *et al.*, Phys. Rev. Lett. **77**, 3343 (1996).
[4] T. Ditmire *et al.*, Nature (London) **386**, 54 (1997).
[5] M. Lezius, S. Dobosz, D. Normand, and M. Schmidt, Phys. Rev. Lett. **80**, 261 (1998).
[6] T. Ditmire *et al.*, Phys. Rev. A **53**, 3379 (1996).
[7] A. V. Gurevich, L. V. Pariiskaya, and L. P. Pitaevskii, Zh. Eksp. Teor. Fiz. 63, 516 (1973) [Sov. Phys. JETP **36**, 274 (1973)].
[8] T. Ditmire *et al.*, Nature (London) **398**, 489 (1999).
[9] J. Purnell, E. M. Snyder, S. Wei, and A. W. Castleman, Jr.,

Chem. Phys. Lett. **229**, 333 (1994).
[10] D. J. Fraser and M. H. R. Hutchinson, J. Mod. Opt. **43**, 1055 (1996).
[11] T. Ditmire *et al.*, Phys. Rev. A **57**, 369 (1998).
[12] R. A. Smith, T. Ditmire, and J. W. G. Tisch, Rev. Sci. Instrum. **69**, 3798 (1998).
[13] O. F. Hagena, Z. Phys. D **4**, 291 (1987).
[14] J. Collier *et al.*, Laser Part. Beams **14**, 283 (1996).
[15] P. E. Young, M. E. Foord, A. V. Maximov, and W. Rozmurs, Phys. Rev. Lett. **77**, 1278 (1996).
[16] G. Manfredi, S. Mola, and M. R. Feix, Phys. Fluids B **5**, 388 (1993).