Explosion of atomic clusters irradiated by high-intensity laser pulses: Scaling of ion energies with cluster and laser parameters

E. Springate,* N. Hay, J. W. G. Tisch, M. B. Mason, T. Ditmire,† M. H. R. Hutchinson, and J. P. Marangos
Blackett Laboratory Laser Consortium, Imperial College of Science, Technology and Medicine, Prince Consort Road, London SW7 2BZ, United Kingdom

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Experimental measurements of scaling with cluster and laser parameters of the energies of ions produced in the explosion of atomic clusters have been obtained, with a view to experimental optimization of the cluster explosion temperature. The noble-gas clusters were irradiated with high-intensity, 200-fs laser pulses. Ion energy scalings with cluster size (ranging from 10^2-10^5 atoms per cluster), laser intensity ($10^{14}-10^{16}\,\mathrm{W\,cm^{-2}}$), and laser wavelength (780 and 390 nm) are presented for both Xe and Kr clusters. Numerical calculations of the interaction, treating the cluster as a spherical nanoplasma, are also presented.

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I. INTRODUCTION

The interaction of clusters with high-intensity laser pulses has been studied by a number of groups over the past few years. Extremely energetic ions with energies up to 1 MeV [1] and keV electrons [2] are produced as noble-gas clusters explode. The ions are stripped to very high charge states [1,3,4] and the x-ray yield is comparable to that from solid targets [5]. Highly stripped ions have also been observed from laser-heated metallic clusters [6]. The observation of neutrons from deuterium clusters irradiated with only ~100 mJ of laser energy [7] demonstrated that nuclear fusion can occur in the plasma formed after the clusters explode.

Several different theories have been developed to explain these observations. McPherson *et al.* [8] described the laser-cluster interaction using a model where collisional ionization can produce inner-shell vacancies in ions. This model was later extended to include coherent electron motion, where the electrons oscillate collectively in the cluster [9]. The ''ionization ignition'' model [10] predicts an avalanche of ionization in a cluster due to the combined field of the laser and cluster ions, leading to a Coulomb explosion of the cluster. A simple Coulomb explosion model [11], tracing the trajectory of a single electron oscillating in the combined field of the laser pulse and a 1100-atom Xe cluster with a mean charge per ion between 1+ and 12+, has indicated that all free electrons can leave the cluster during one optical cycle. In contrast, classical particle dynamics simulations [12] show that, even for clusters as small as 55 Ar atoms, a substantial fraction of ionized electrons can be confined to the bulk of the cluster by space charge. If the electrons are retained in the cluster, it is then valid to treat the cluster as a plasma.

The model developed by Ditmire *et al.* [5], which treats the expanding cluster as a spherical nanoplasma, suggests that the cluster explosion is driven by an enhancement in the electron heating in the cluster. This enhancement occurs when the electron density in the cluster drops to three times the critical electron density, n_{crit} (the electron density at which the laser frequency equals the plasma frequency, given by $n_{\mathrm{crit}}=\pi c^2 m_e/e^2\lambda^2$). At this point the electric field in the cluster is greater than the vacuum electric field, there is a peak in the instantaneous electron temperature, and the cluster expansion velocity increases dramatically. The model also predicts a characteristic double-peaked electron spectrum, which has been observed experimentally [2]. The sharp ''hot electron'' peak in the spectrum is due to electrons which leave the cluster at the $3n_{\mathrm{crit}}$ resonance. The existence of an optimum pulse width for a given cluster size [13] is also evidence of this resonance in heating, as the cluster size determines the time taken for the electron density in the cluster to drop to $3n_{\mathrm{crit}}$ and this point has to be near the peak of the laser pulse for optimal heating.

Experimental optimization of the cluster explosion temperature through the adjustment of laser and cluster parameters may, for example, increase the yield of fusion neutrons to provide a source of an ultrafast pulse of monoenergetic neutrons [7], boost the x-ray yield [14], and enable the production of highly stripped ions for possible x-ray laser schemes. In this paper we present measurements of the scalings of the ion energies produced from laser-irradiated noble-gas clusters as a function of cluster size (10^2-10^5 atoms per cluster), laser intensity ($10^{14}-10^{16}\,\mathrm{W\,cm^{-2}}$), and laser wavelength (780 and 390 nm) for two different cluster ion species (Xe and Kr).

Our experimental measurements have been compared with numerical calculations to provide a more stringent test of the nanoplasma model [5]. We find that our experimental measurements of the scaling of the ion energies with cluster size and laser intensity show good agreement with the predictions of the nanoplasma model. In particular, the existence of an optimum cluster size for a given laser pulse provides strong evidence for resonant heating of the cluster plasma. However, the nanoplasma model predicts a stronger scaling with laser wavelength than is observed experimentally.

*Present address: FOM-Institute for Atomic and Molecular Physics, Kruislaan 407, 1098 SJ Amsterdam, The Netherlands.

†Present address: the Laser Program, L-477, Lawrence Livermore National Laboratory, Livermore, California 94550.

II. EXPERIMENTAL DETAILS

The measurement of ion energies is described in detail in Ref. [15]. In our experiment a beam of atomic clusters, produced in the expansion of a high-pressure gas into vacuum, was irradiated by a focused, high-intensity, femtosecond laser beam. The electrons and ions expelled from the clusters with velocities perpendicular to both the cluster beam and the laser beam propagated along a 38-cm flight tube and were detected by a microchannel plate detector (MCP). The input face of the MCP was held at -2 kV, but a grounded metal grid placed ~ 2 mm before the MCP ensured that the flight tube was field-free. The ion energies were then determined by time-of-flight measurements in the drift tube. The presence of the electric field between the grid and MCP had a negligible effect on the flight times from which the ion kinetic energies were calculated. The field also prevented electrons with energies less than 2 keV from reaching the detector.

The laser used was a Ti sapphire chirped pulse amplification laser (described in Ref. [16]), which delivered ~ 200 fs pulses at a wavelength of 780 nm. The pulse length ranged from 170 to 230 fs during the experiments described in this paper. The laser was focused at $f/10$ to a peak focused intensity of $2\times10^{16}\,\mathrm{W\,cm^{-2}}$ with 30 mJ of laser energy. This intensity was confirmed in a separate experiment by observing the appearance of Xe^{+} to Xe^{3+} through over-the-barrier ionization. The laser intensity was controlled using a half-wave plate and polarizer before the final amplifier. Prepulse on a nanosecond time scale was measured to be less than 3×10^{-6} of the main pulse. The laser was horizontally polarized, so the electric field of the fundamental was oriented along the axis of the detector. For some experiments, the laser was frequency doubled using a 3-mm type-1 KDP crystal, which meant that the second-harmonic radiation was vertically polarized. The maximum second-harmonic energy was 3 mJ, which could be focused to a peak intensity of $1\times10^{16}\,\mathrm{W\,cm^{-2}}$ (this was again confirmed through over-the-barrier ionization of Xe).

The extent of clustering in the gas jet was estimated using Hagena's scaling parameter [17], and the presence of clusters in the jet was confirmed through Rayleigh scattering measurements [18]. Rayleigh scattering measurements can give no information on the distribution of cluster sizes produced. However, measurements of the size distribution of large noble-gas clusters [19,20] have suggested that the cluster sizes have a log-normal distribution with a full width at half maximum approximately equal to the mean cluster size. The error on our estimate of the mean cluster size is of the order of a factor of 2, corresponding to an error of $\sim 25\%$ on the cluster radius. In the experiments described here, the mean cluster size was controlled by changing the backing pressure of the gas jet. Our Rayleigh scattering measurements indicate that the mean cluster size varies as $(\text{backing pressure})^{2.4\pm0.1}$ [21].

A skimmer was used to collimate the cluster beam, giving a low-average-density cluster beam ($\sim 10^{14}\,\mathrm{atoms\,cm^{-3}}$) that intercepted the laser beam at the focus. Electron and ion signals were only observed when the laser pulse was coincident with the arrival of clusters from the gas jet.

The experimental results have been compared with numerical calculations based on the nanoplasma model of the laser-cluster interaction [5]. The numerical calculations presented throughout this paper represent the interaction of a laser pulse with a single cluster. They have not been averaged over the Gaussian intensity distribution in the focus or the cluster size distribution. The cluster size quoted throughout is the mean of the experimental distribution. The cluster size and laser intensity distributions will be important experimentally, so the numerical calculations cannot be expected to match the experimental data exactly. They can, however, reproduce the trends in the observed scalings. The maximum ion energy is given directly by the model from the expansion velocity of the cluster.

The low number density of clusters (due to the presence of the skimmer) means that we study the interaction of the laser with isolated clusters rather than the bulk plasma formed after the cluster has expanded. Measurements of absorption and x-ray yield are typically performed directly below the gas jet, where the number density of clusters is $\sim 10^{5}$ higher than in the experiments described in this paper.

III. RESULTS

A raw time-of-flight (TOF) spectrum (obtained by averaging over 200 laser shots) from 14 000-atom Xe clusters irradiated by a 780-nm pulse with a peak intensity of $1\times10^{16}\,\mathrm{W\,cm^{-2}}$ and the ion energy spectrum obtained from it are shown in Fig. 1. The spike in the time-of-flight spectrum near zero time is due to electrons with energies above 2 keV, while the lobe extending out from ~ 500 ns is due to ions. With xenon in a 38-cm drift tube, 1-MeV ions have a flight time of 300 ns, and 100-keV ions have a 1-μs flight time. This spectrum has a mean ion energy $\overline{E}$, defined as

$$\overline{E}=\frac{\int_{E_{\min}}^{E_{\max}} E f(E)\,dE}{\int_{E_{\min}}^{E_{\max}} f(E)\,dE}, \tag{1}$$

of 42 keV and a maximum of 350 keV.

The maximum ion energy is extracted from the raw TOF traces using a numerical algorithm to find the earliest ion hits. The algorithm integrates the TOF spectrum and then fits a straight line to the noise between the electron peak and the ion peak. The point where the signal starts to increase above the noise defines the shortest flight time and maximum ion energy. The error on the maximum ion energy is the error on the shortest flight time, combined with the error on the position of zero time (defined as the time at which the center of the laser pulse reaches the focus).

Figure 2 shows the mean ion energy as a function of angle with respect to the direction of laser polarization. The polarization was rotated by placing a half-wave plate in the beam path just before the TOF chamber. The mean ion energy is $\sim$ 15% higher along the direction of laser polarization than perpendicular to it. Some anisotropy of the exploding ions is

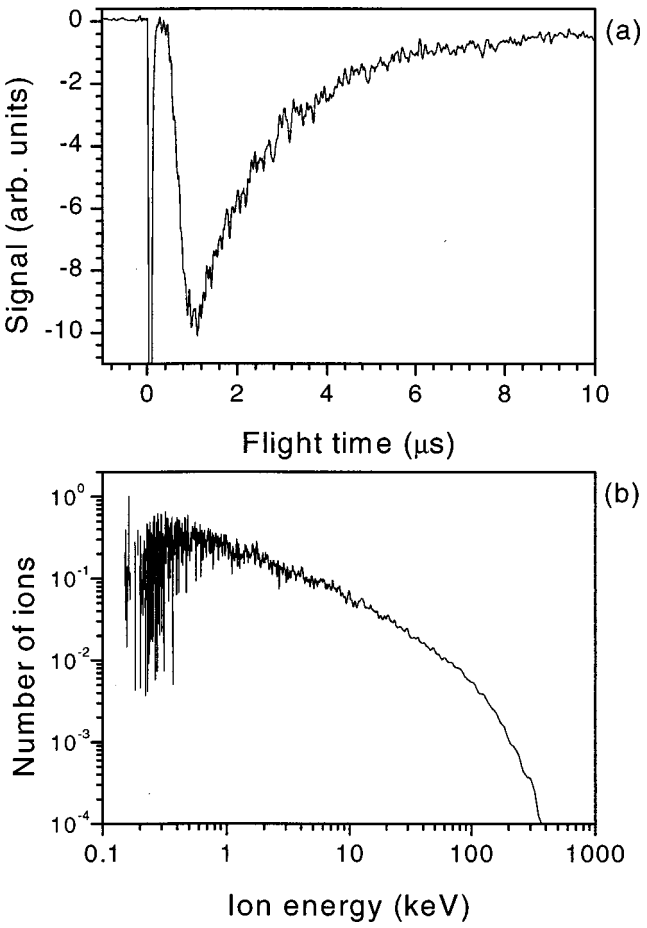

FIG. 1. (a) Time-of-flight spectrum from 14 000-atom Xe clusters irradiated by a peak intensity of $1\times10^{16}\,\mathrm{W\,cm^{-2}}$. The 38-cm flight tube was field-free. (b) Ion energy spectrum from clusters of 9000 Xe atoms irradiated by a peak intensity of $1.5\times10^{16}\,\mathrm{W\,cm^{-2}}$, derived from the time-of-flight trace in (a). The mean ion energy is 42 keV and the maximum is 350 keV.

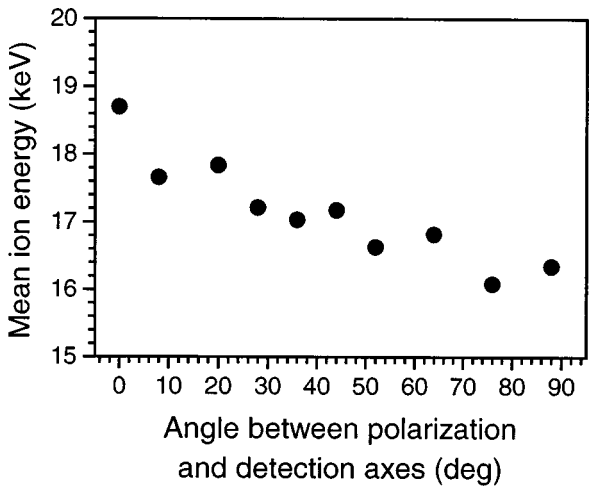

FIG. 2. Measured mean ion energies as a function of the angle between the laser polarization and the detection axis. The 5300-atom Xe clusters were irradiated with 170 fs, 780-nm laser pulses with a peak intensity of $1.3\times10^{16}\,\mathrm{W\,cm^{-2}}$.

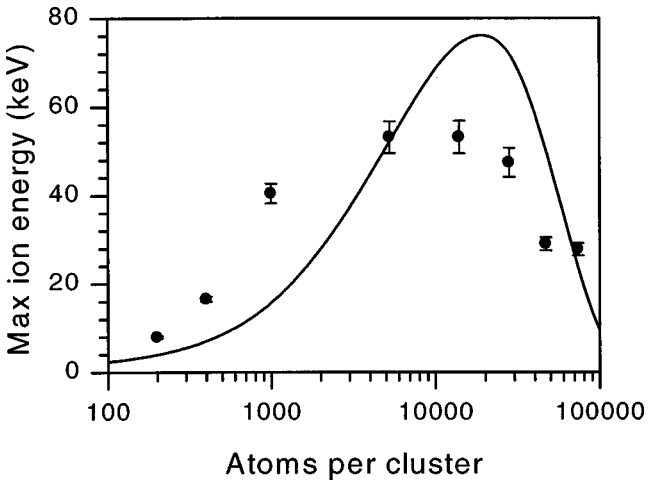

FIG. 3. Measured (solid circles) and calculated (solid line) maximum ion energies as a function of cluster size for clusters irradiated with a 230 fs, 780-nm laser pulses with a peak intensity of $3\times10^{15}\,\mathrm{W\,cm^{-2}}$.

to be expected as the ''warm'' electrons (those with energies in the range 0.3–2 keV) are also emitted anisotropically, with a peak along the direction of the laser polarization [2].

A. Scaling with cluster size

Figure 3 shows the variation in maximum ion energies with cluster size for clusters of 200 to 74 000 Xe atoms irradiated with a laser pulse of $3\times10^{15}\,\mathrm{W\,cm^{-2}}$. Fast ions (with energies above 1 keV) were first observed in the time-of-flight signal at a backing pressure of 1 to 1.3 bar. This pressure corresponds to the onset of hot electron production from the exploding clusters [2]. The time-of-flight spectra also show that electrons with energies above 2 keV were only detected when hot ions are produced. This points to a change in the dynamics of the cluster expansion once the cluster size increases to above ~200–400 atoms.

The maximum ion energy rises from 8 keV at 1 bar (200 atoms/cluster) to a peak of 53 keV at clusters of ~10 000 ±5000 atoms before falling to 28 keV as the cluster size is increased beyond 50 000 atoms/cluster. Numerical calculations from the nanoplasma model of the maximum ion energy as a function of cluster size are also shown (solid line). The model predicts the existence of an optimum cluster size of ~20 000 atoms. The predicted variation in ion energy with cluster size is in good agreement with that measured experimentally.

The nanoplasma model suggests that the dynamics of the explosion is governed by the point in the laser pulse at which the cluster experiences the resonant heating. The highest ion temperatures are obtained when the cluster passes through the $3n_{\mathrm{crit}}$ point close to the peak of the laser pulse. Small clusters expand more quickly and reach this point before the peak: larger clusters pass through $3n_{\mathrm{crit}}$ well after the peak. Increasing the intensity increases the initial ionization and expansion rates so that the $3n_{\mathrm{crit}}$ point occurs earlier in the laser pulse for a given cluster size, thus shifting the optimum size towards larger clusters. The existence of an optimum

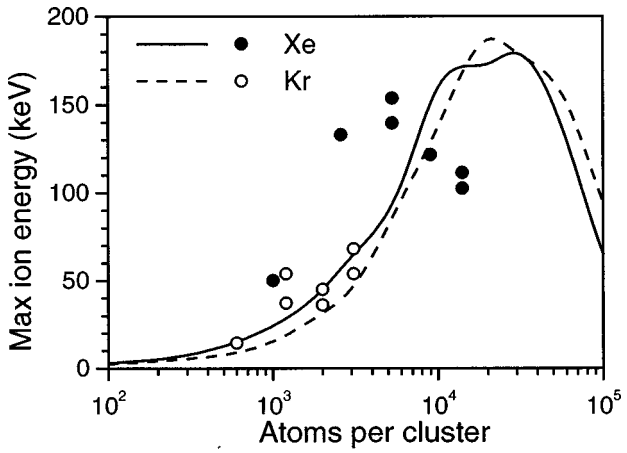

FIG. 4. Measured (circles) and calculated (lines) maximum ion energies from Xe (filled circles, solid lines) and Kr (open circles, dotted lines) clusters with a variety of cluster sizes. The laser intensity was $1.3\times10^{16}\,\mathrm{W\,cm^{-2}}$ and the pulse length was 170 fs.

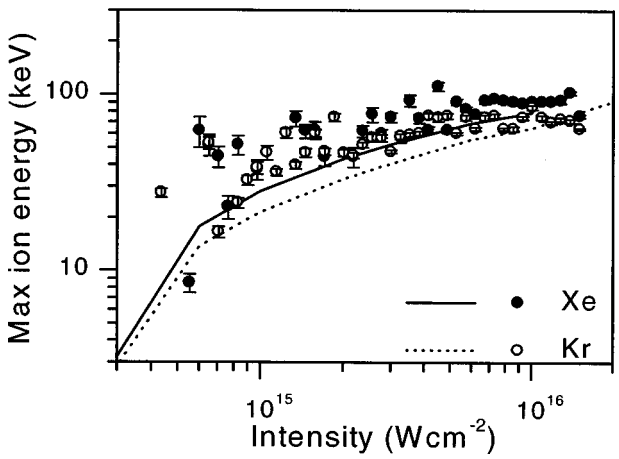

FIG. 5. Maximum ion energies measured as a function of laser intensity for 5300-atom Xe clusters (filled circles) and 6200-atom Kr clusters (open circles), together with numerical calculations for Xe clusters (solid lines) or Kr clusters (dotted lines). The 780-nm laser pulse was 230-fs long.

cluster size for a given set of laser-pulse parameters demonstrated here, together with the existence of an optimum laser-pulse width for a given cluster size [13], is strong evidence for a resonance in the cluster heating.

With Kr clusters, we observe similar behavior to that of Xe clusters, but the ion energies are slightly lower. Figure 4 shows the maximum ion energies from Xe and Kr clusters as a function of cluster size, together with numerical calculations. This set of experiments (a different dataset to Fig. 3) was carried out with a 170-fs laser pulse with a peak intensity of $1.3\times10^{16}\,\mathrm{W\,cm^{-2}}$. At low cluster sizes ($<10\,000$ atoms) the mean ion energies from Xe clusters are ~5-keV higher than those from Kr clusters. The condensation parameter for Kr is 2900 compared to 5500 for Xe [20], meaning that the clusters are approximately half the size for the same gas-jet backing pressure. This prevented us from increasing the cluster size to the point where an optimum cluster size could be observed. However, over the region of cluster sizes for which we have data, the trends in the data are consistent with the model.

B. Intensity dependence

We have also examined the scaling of the ion energies with laser intensity in both Xe and Kr clusters (Fig. 5). The Xe clusters contained ~5300 atoms (corresponding to a 43-Å radius) and the krypton clusters ~6200 atoms (41 Å). Again, similar behavior is observed for both ion species, with ~20% lower energies for Kr.

In Xe our measurements show a sharp onset of hot ion production at an intensity of $6\times10^{14}\,\mathrm{W\,cm^{-2}}$. The ion energies rise steeply up to $\sim1\times10^{15}\,\mathrm{W\,cm^{-2}}$, where the maximum ion energy is ~50 keV. At intensities higher than these, the maximum ion energy increases only as $\sim I^{0.2}$ with increasing intensity while the integrated ion yield increases as $\sim I^{1.4}$, consistent with the increase in focal volume (an $I^{3/2}$ scaling would be expected for a Gaussian focal spot). At $1\times10^{16}\,\mathrm{W\,cm^{-2}}$, the maximum ion energy is 90 keV. The krypton ion energies follow a similar trend to the xenon, with a sharp increase up to a mean energy of 9 keV at $10^{15}\,\mathrm{W\,cm^{-2}}$ followed by a slow increase up to energies of 75 keV at $10^{16}\,\mathrm{W\,cm^{-2}}$.

The calculated intensity dependence of the mean ion energy attained in the explosion is also shown in Fig. 5. The nanoplasma model predicts that xenon and krypton will exhibit very similar scalings with intensity with the laser and cluster parameters used in the experiment. For both ion species, the ion energy increases sharply with laser intensity from 40 eV at $6\times10^{13}\,\mathrm{W\,cm^{-2}}$ to 2 keV at $3\times10^{14}\,\mathrm{W\,cm^{-2}}$. In this intensity region, the amount of ionization is sufficient that the electron density within the cluster reaches $3n_{\mathrm{crit}}$ at which point the field in the cluster is enhanced with respect to the laser field and the cluster expansion velocity increases. The expansion velocity is not, however, large enough compared to the ionization rate that the electron density in the cluster can drop to $3n_{\mathrm{crit}}$ again before the laser pulse has passed.

When the laser intensity is greater than $\sim6\times10^{14}\,\mathrm{W\,cm^{-2}}$ the cluster is ionized enough to reach $3n_{\mathrm{crit}}$ fairly early on in the laser pulse. It then starts to expand and the combination of collisional ionization and cluster expansion causes the electron density to rise and then fall through $3n_{\mathrm{crit}}$ before the laser intensity drops off. As the electron density drops through $3n_{\mathrm{crit}}$ for the second time, the field in the cluster is strongly enhanced and the radial acceleration increases dramatically. In this intensity regime the ion energies increase only slowly with increasing intensity (as $\sim I^{0.5}$) because as the laser intensity increases, the amount of initial ionization increases. This means that the cluster expands faster and the electron density drops through $3n_{\mathrm{crit}}$ earlier in the laser pulse. The cluster does not then experience a substantially higher laser intensity at the point of resonant heating.

The numerical results are in extremely good agreement with the experimental measurements, matching the general trends and predicting the intensity regime where the ion energies increase most rapidly. Although the scaling of ion

energy with laser intensity is fairly weak from 10^{15}–$10^{16}\,\mathrm{W\,cm^{-2}}$, the model predicts that the ion energies will start to rise more steeply at intensities above $3\times10^{16}\,\mathrm{W\,cm^{-2}}$. However, this is a higher intensity than is currently accessible in our experiments.

C. Wavelength scaling

We have examined the wavelength scaling of the cluster explosion dynamics by carrying out a comparative study of the ion energy spectra at the laser fundamental and the second harmonic. For these experiments the laser was frequency doubled to 390 nm using a 3-mm-thick type-1 KDP crystal. The second harmonic was vertically polarized for all these experiments which means that the electric-field vector was perpendicular to the detection axis.

Similar behavior is found in the cluster explosion with IR (780 nm) and blue (390 nm) light. As with IR irradiation, the mean and maximum ion energies with 390-nm light are approximately 15% lower perpendicular to the laser polarization than parallel to it. The ion energies in the blue presented here are measured perpendicular to the laser polarization and have not been scaled up for comparison with the IR. Numerical calculations comparing the ion energies from clusters irradiated with the two wavelengths are also presented and it is apparent that there is some disagreement between experiment and theory.

The maximum ion energies resulting from the irradiation of Xe clusters varying in size from 200 to 74 000 atoms irradiated by $5\times10^{15}\,\mathrm{W\,cm^{-2}}$ of 390-nm radiation, together with those from clusters irradiated by $3\times10^{15}\,\mathrm{W\,cm^{-2}}$ at 780 nm (from Fig. 3) are shown in Fig. 6. Experimentally, we find that hot ions are only produced with backing pressures greater than one bar, when clusters of more than 200 atoms are present in the jet, as was found with IR irradiation. The maximum ion energy rises to a peak of ~35 keV at the optimum cluster size of 28 000 atoms and decreases as the cluster size increases beyond this point. The ion energies from clusters irradiated at 390 nm are substantially lower than those from 780-nm irradiation, except possibly at the largest cluster sizes.

In contrast, the nanoplasma model predicts that at cluster sizes below ~10 000 atoms, the mean energy in the blue is ~5-keV higher with 390-nm irradiation than with 780 nm. The ion energies obtained with the second harmonic are predicted to be much higher than with the fundamental for large cluster sizes. The optimum cluster size is calculated to be ~20 000 atoms at 780 nm and 80 000 atoms at 390 nm. Both experiment and model agree that the optimum cluster size is larger in the blue than the IR.

Experimental measurements of the intensity dependence of the mean and maximum ion energies in the IR and the blue for clusters of 5300 Xe atoms show that the intensity scaling (Fig. 7) is quite different for the two wavelengths. With 390-nm irradiation, the maximum ion energies increase as $\sim I^{1.1}$ in the intensity range $8\times10^{14}\,\mathrm{W\,cm^{-2}}$ to $5\times10^{15}\,\mathrm{W\,cm^{-2}}$, while with 780-nm irradiation they increase only as $I^{0.2}$ in this intensity region. The same scalings were also observed with 6200-atom Kr clusters, but again the ion energies were slightly lower.

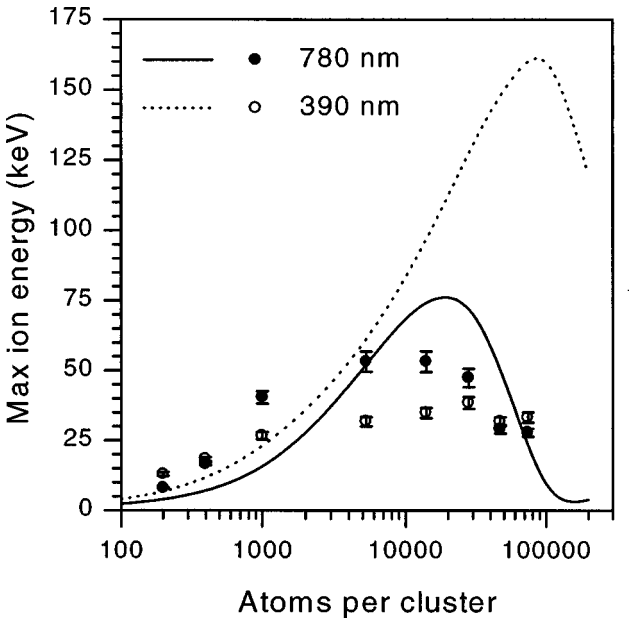

FIG. 6. Measured (circles) and calculated (lines) maximum ion energies measured in the interaction of xenon clusters with 780-nm (filled circles, solid lines) and 390-nm (open circles, dotted lines) radiation. The peak intensity of the 390-nm pulses was $5.0\times10^{15}\,\mathrm{W\,cm^{-2}}$ and that of the 780-nm pulses was $2.8\times10^{15}\,\mathrm{W\,cm^{-2}}$. The calculation assumes a 230 fs, 780-nm pulse or a 160-fs, 390-nm pulse, both having peak intensities of $3\times10^{15}\,\mathrm{W\,cm^{-2}}$.

The nanoplasma model predicts that the scaling of ion energy with increasing laser intensity is very similar at both wavelengths, as shown in Fig. 7. With both wavelengths, the model predicts a sharp increase followed by a plateau where the maximum ion energy is $\sim5\times10^4$ eV. The onset of the

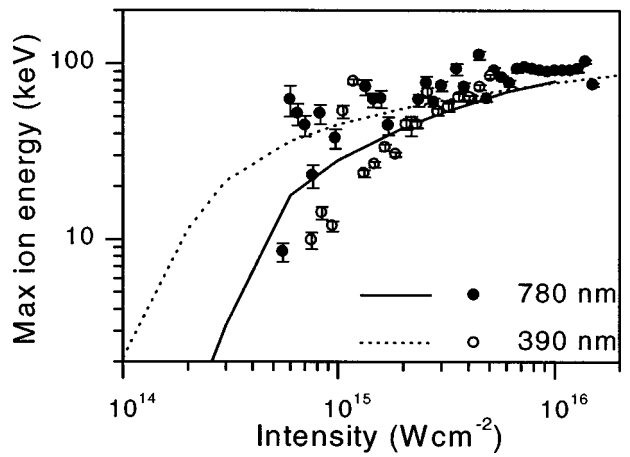

FIG. 7. Measured (circles) and calculated (lines) intensity dependence of the maximum ion energies from the irradiation of 5300-atom Xe clusters with 390-nm (open circles, dotted line) and 780-nm (filled circles, solid line) laser pulses.

plateau is at a lower intensity with the blue–at $2\times10^{14}\,\mathrm{W\,cm^{-2}}$ compared to $6\times10^{14}\,\mathrm{W\,cm^{-2}}$ in the IR. This is because the increased electron collisional heating rate with 390-nm irradiation leads to a higher thermal ionization rate. This in turn means that the initial (relatively slow) expansion of the cluster is more rapid, so the electron density drops through the $3n_{\mathrm{crit}}$ explosion point earlier in the laser pulse. At intensities from $2-6\times10^{14}\,\mathrm{W\,cm^{-2}}$ a cluster irradiated by a 390-nm pulse will pass through the $3n_{\mathrm{crit}}$ resonant heating point before the laser pulse passes, but a cluster irradiated at 780 nm will not.

The difference between the experimental intensity scalings at the two wavelengths may in part be due to the different pulse shapes of the laser fundamental and second harmonic. Any prepulse and pedestal on the fundamental is strongly reduced in frequency doubling as, unless the crystal is saturated, the second-harmonic pulse shape follows the square of the fundamental intensity. However, a long tail can be introduced on the second-harmonic pulse due to the group velocity mismatch between the two wavelengths ($83\,\mathrm{fs\,mm^{-1}}$ for 780-nm pulses frequency doubled in KDP) [22].

Another possible reason for the difference in behavior between the two wavelengths is the difference in the multiphoton ionization (MPI) rates. MPI is expected to occur at low intensities on the leading edge of the laser pulse and, at a given intensity, we will be further ''in'' the multiphoton regime with shorter-wavelength radiation. However, inclusion of MPI rates in the numerical model showed that the initial ionization rate has only a weak influence on the final ion temperature, the explosion being dominated by collisional ionization once a few electrons have been freed.

IV. DISCUSSION

We have studied how the energies of ions produced in the explosion of atomic clusters depend on cluster size (10^2-10^5 atoms per cluster), laser intensity ($10^{14}-10^{16}\,\mathrm{W\,cm^{-2}}$), and laser wavelength (780 and 390 nm) for two cluster ion species (Xe and Kr). Our investigation of the scaling of ion energies with cluster size shows that the mean and maximum ion energies increase as the cluster size increases up to an optimum cluster size, from which point they decrease. This observation is in agreement with the results of Zweiback *et al.* [13] who have examined the relationship between cluster size and laser-pulse length for optimal cluster heating.

The nanoplasma model suggests that the dynamics of the cluster explosion is governed by the time in the laser pulse at which the electron density in the cluster falls through $3n_{\mathrm{crit}}$, where the electron heating rate is dramatically enhanced. The highest ion temperatures are obtained when the cluster experiences this resonant heating close to the peak of the laser pulse. This explains the existence of an optimum cluster size for a given pulse length, and an optimum pulsewidth for a given cluster size. Clusters smaller than the optimum size expand more quickly and reach the $3n_{\mathrm{crit}}$ point before the peak of the laser pulse, while if the clusters are larger than the optimum they pass through this point well after the peak. If the pulse width is too short for the cluster size, $3n_{\mathrm{crit}}$ is on the rising edge of the laser pulse; too long and $3n_{\mathrm{crit}}$ is on the falling edge.

As well as enabling the optimization of cluster heating, the existence of these optimum cluster sizes and pulse lengths provides support for the nanoplasma model. Coulomb explosion models predict that the ion energies increase monotonically as the cluster size increases [23]. Studies of the ionization ignition mechanism indicate that the lowering of potential barriers due to the close presence of other charged ions, which leads to enhanced ionization rates, increases as the number of ions surrounding each ion increases [24]. However, for larger clusters the ionization ignition model predicts little dependence on the degree of clustering as only the closest ions efficiently lower the ionization potential of the ions within a cluster. The inner-shell excitation and coherent electron motion models [8,9] predict that, for a given intensity, there will be an allowed zone of cluster sizes for which x-ray emission from a given shell will occur, with the number of x rays produced scaling as $(\mathrm{cluster\ size})^{4/3}$.

The effect of the laser intensity on the cluster explosion dynamics has also been examined. We observed a sharp onset of hot ion production at a laser intensity of $6\times10^{14}\,\mathrm{W\,cm^{-2}}$. The mean and maximum ion energies increase rapidly as the laser intensity is increased to $1\times10^{15}\,\mathrm{W\,cm^{-2}}$, from which point the ion energies increase only slowly. The integrated ion yield scales as $I^{1.4}$ for intensities above $1\times10^{15}\,\mathrm{W\,cm^{-2}}$, consistent with the increase in focal volume. This result is in good agreement with the numerical calculations based on the nanoplasma model. The weak dependence of the cluster explosion dynamics on laser intensity at higher intensities has also been seen by Dobosz and co-workers [25]. They performed x-ray spectroscopy of cluster nanoplasmas, calculated the mean number of x-ray photons emitted per laser pulse and found that it scales as $I^{3/2}$, again consistent with the increase in focal volume. For Kr clusters of 7×10^5 atoms, the explosion process was found to saturate below $4\times10^{16}\,\mathrm{W\,cm^{-2}}$.

We have used the first and second harmonics of the Ti: sapphire laser (at 780 and 390 nm) to examine the wavelength scaling of the cluster explosion dynamics. We find that similar behavior is found in the cluster explosion at both wavelengths. However, the nanoplasma model is unable to reproduce successfully the change in the variation of the ion energies with cluster size and laser intensity. With 390 nm, there is an optimum cluster size for a given laser pulse and, as predicted by the nanoplasma model, the optimum cluster size is larger in the blue than the IR. However, the measured ion energies with 390-nm radiation are lower than with 780-nm radiation over a wide range of cluster sizes and laser intensities, contrary to the prediction of the nanoplasma model. We find that the intensity dependence of the ion energies at 390 nm is different to that at 780 nm, scaling as $I^{1.1}$ in the blue rather than the sharp increase followed by a leveling off as found for the IR and as predicted by the model. The difference in intensity scalings could, however, be partly due to the difference in pulse shapes between the fundamental and second harmonic.

The wavelength scaling of cluster dynamics has been examined by several groups. Kondo *et al.* [26,27] found that

the x-ray yield from clusters was much lower at 800 nm than 248 nm. In contrast to these results, Dobosz *et al.* [25] found that there was no strong scaling of x-ray yield from first to second harmonic. Our numerical calculations with the nanoplasma model indicate that there is no substantial change in the time history of the cluster dynamics when the wavelength of the radiation used to heat the cluster is varied from 250 nm to 1μm.

The nanoplasma model provides a good prediction of the heating of the cluster plasma, enabling the experimental optimization of the heating through adjustment of the cluster and laser parameters. However, the model assumes the isotropic expansion of a uniform sphere. With a uniform isotropic expansion, the ion energy increases linearly from zero with the cluster radius, so the number of ions at each velocity ν will be proportional to ν^2. This does not correspond to the measured spectrum. The nanoplasma model is inadequate for modeling the expansion of the cluster plasma after it is heated. Ditmire *et al.* [15] found the self-similar solution of the ion fluid equations for an isotropic, radial expansion for a Xe plasma with a mean charge of 20+ and an electron temperature of 2.5 keV and found that the calculated ion energy spectrum was very close to that obtained from a 2500-atom Xe cluster, suggesting that the expansion is driven largely by hydrodynamic pressure.

V. CONCLUSIONS

Our measurements of the ion energies obtained from the explosion of clusters irradiated by intense, femtosecond laser pulses are in good agreement with the heating mechanisms of the nanoplasma model developed by Ditmire *et al.* [5]. Experimental measurements of the ion energies as a function of laser and cluster parameters allow the optimization of the cluster explosion temperature. In particular, the existence of an optimum cluster size both allows the cluster heating to be maximized for a given laser pulse and provides strong support for the resonant heating of the cluster plasma. This has important implications for potential applications such as nuclear fusion in clusters [7] and the production of short-pulse x rays [14]. Further enhancement of the ion energies may be possible through multiple-pulse heating schemes [13,28].

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