Enhanced explosion of atomic clusters irradiated by a sequence of two high-intensity laser pulses

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Experiment and numerical simulations have shown that the heating of atomic clusters can be enhanced by using a correctly timed sequence of two high-intensity pulses of different frequencies. We have measured ion energies from Xe clusters irradiated with two high-intensity femtosecond laser pulses, one at the laser fundamental frequency (780 nm) and one at its second harmonic (390 nm), focused to an intensity of $\sim 10^{15}$ W cm^{-2}. The ion energies are found to depend on the relative delay of the two pulses but not on their relative polarizations. For the optimum value of the delay between the two laser pulses, the maximum ion energy is doubled from 100 to 200 keV. The laser energy required to obtain a given ion energy is significantly less when the cluster is irradiated with two pulses of different frequencies than a single laser pulse.

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The interaction of clusters with high-intensity laser pulses has been studied by a number of groups in the past few years. Clusters form an intermediate state between atoms or small molecules and solids, having the same local density as a solid but a radius much smaller than a laser wavelength. Extremely energetic ions, with energies up to 1 MeV [1], and keV electrons [2] are produced as the atomic 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]. Irradiation of clusters leads to far more energetic particles than irradiation of single atoms or small molecules at the same intensity.

A 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}), at which 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 numerical model is in good qualitative agreement with our experimental results, reproducing trends in ion energy with cluster size, laser intensity [6], and cluster ion species [7]. 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 cluster size for a given pulse width [6] and an optimum pulse width for a given cluster size [8] are 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.

The model suggests that the explosion dynamics can be manipulated by irradiating the cluster with two laser pulses of different frequencies in sequence. The critical electron density is wavelength dependent ($n_{\mathrm{crit}}=\pi c^2 m_e/e^2\lambda^2$), so the $3n_{\mathrm{crit}}$ enhanced heating point will occur at a higher electron density for shorter wavelengths. This suggests that a cluster irradiated with two laser pulses of different wavelengths will experience enhanced heating twice in its expansion.

In this paper, we present experimental results and numerical simulations which show a change in the cluster dynamics when the cluster is irradiated by two pulses of different frequencies in sequence. The cluster experiences two peaks in its heating and electron temperature, which leads to an increase in the energies of the ions produced in the explosion. We present experimental measurements of ion energies from Xe clusters irradiated with high-intensity femtosecond laser pulses, at the laser fundamental frequency (780 nm) and its second harmonic. The mean and maximum ion energies are found to depend on the relative delay of the two pulses but not on their relative polarizations. The mean ion energy is doubled for the optimum value of the delay between the two laser pulses, with good agreement between the observed behavior and the predictions of the model.

The nanoplasma model of the laser cluster interaction [5] has been adapted to simulate the irradiation of a cluster by two laser pulses with arbitrary frequencies, intensities, and relative delays. In vacuum, the two-color field of frequencies ω_1 and ω_2 is given by

$$E_0 = E_{01}e^{i\omega_1 t} + E_{02}e^{i(\omega_2 t+\varphi)} + \text{c.c.} \tag{1}$$

The field inside the cluster is then calculated from the external field by [from Eq. (16) in Ref. [5]]

$$E = E_1 + E_2 = \frac{3}{|\varepsilon_1+2|}E_{01}e^{i\omega_1 t} + \frac{3}{|\varepsilon_2+2|}E_{02}e^{i\omega_2 t} + \text{c.c.} \tag{2}$$

The dielectric constants $\varepsilon_{1,2}$ [Eq. (18) in Ref. [5]], which are a function of optical frequency, plasma frequency, and collision frequency, are calculated for each laser wavelength, as are the collision frequencies [Eqs. (20) and (21) in Ref. [5]]. The heating is assumed to be through collisional heating of

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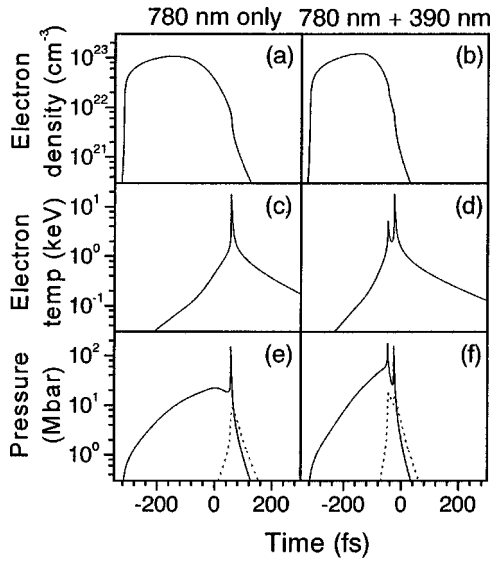

FIG. 1. Numerical modeling of the interaction of a cluster of 14 000 Xe atoms with a single high-intensity laser pulse (780 nm, 260 fs, 4×10^{15} W cm^{-2}) (a), (c), and (e) or with two laser pulses of different frequencies (780 nm, 260 fs, 4×10^{15} W cm^{-2} and 390 nm, 185 fs, 2.5×10^{15} W cm^{-2}) (b), (d), and (f). The center of each laser pulse is at $t=0$ fs. (a) and (b) show the electron density inside the cluster, (c) and (d) show the electron temperature, and (e) and (f) show the hydrodynamic pressure (solid line) and Coulomb pressure (dotted line).

the electrons by the laser field (inverse bremsstrahlung). The time-averaged energy-deposition rate [Eq. (15) in Ref. [5]] then becomes

$$\frac{\partial U}{\partial t}=\frac{\omega_1}{8\pi}\mathrm{Im}[\varepsilon_1]|E_1|^2+\frac{\omega_2}{8\pi}\mathrm{Im}[\varepsilon_2]|E_2|^2. \qquad (3)$$

The cross terms vanish in the time-averaged heating rate between the two fields: the total heating rate is just the sum of the heating rates due to the two individual fields.

The results of numerical simulations of a xenon cluster irradiated by a 390-nm pulse and a 780-nm pulse are shown in Figs. 1(b), 1(d), and 1(f). Both pulses are Gaussians centered at $t=0$, the 260 fs (full width at half maximum), 780-nm pulse having a peak intensity of 4×10^{15} cm^{-2} and the 185 fs, 390-nm pulse having a peak intensity of 2.5×10^{15} W cm^{-2}. As in the case of a cluster irradiated by a single laser pulse at 780 nm [Figs. 1(a), 1(c) and 1(e)], a few free electrons are tunnel ionized early on in the laser pulse. The electron density rises through $3n_{\mathrm{crit}}$ for both frequencies, at which points the field in the cluster is enhanced and more electrons are liberated through tunnel, laser-driven, and thermal ionization. The electron density is then high enough to shield the field in the cluster from the external laser field. The tunnel and laser-driven ionization rates fall off but electrons are still created through thermal ionization.

The hydrodynamic pressure of the electrons causes the cluster to expand and this, combined with the free streaming of some electrons out of the cluster, means that the electron density starts to drop. At $t=-47$ fs, the electron density [Fig. 1(b)] is 2.2×10^{22} cm^{-3}, three times the critical density for 390-nm radiation. The increased heating rate at $3n_{\mathrm{crit}}$ causes a peak in the electron temperature [Fig. 1(d)] and in the Coulomb and hydrodynamic pressures [Fig. 1(f)] leading to an increase in the radial acceleration and rate at which electrons free stream out of the cluster. After a further 22 fs, at $t=-25$ fs, the electron density is $3n_{\mathrm{crit}}$ for the 780-nm radiation. The cluster again experiences a peak in the electron temperature, Coulomb and hydrodynamic pressure, radial acceleration, and free-streaming rate. The effect of the ''double kick'' from the resonant heating from both fields is to increase the final ion energies to 148 keV (compared to 100 keV from just the IR pulse and 91 keV from the blue pulse alone). When the cluster is irradiated by a single laser pulse, or a sequence of laser pulses with the same frequency, it experiences only one period of resonant heating.

The final ion energies depend on the relative delays of the two pulses. Essentially, the final ion energies are optimized when the $3n_{\mathrm{crit}}$ resonant heating point falls close to the peak intensity of each laser pulse. This condition is satisfied for a four times higher electron density at 390 nm than at 780 nm. As the electron density is falling through the resonant heating points, the optimum heating will occur when the high-frequency pulse precedes the low-frequency pulse.

We have examined the ions produced in the interaction of Xe clusters with a sequence of two high-intensity, femtosecond laser pulses of different frequencies—the laser fundamental and its second harmonic. Our Ti:sapphire chirped pulse amplification laser system produces 780-nm pulses with energies of up to 40 mJ and a pulse length of 255 fs. The second harmonic was produced by frequency doubling in a 3-mm-type I potassium dihydrogen phosphate (KDP) crystal. We assume the second-harmonic pulse duration was $1/\sqrt{2}$ of the IR pulse length (180 fs), as the doubling crystal was not near saturation. The two frequencies were separated by a dichroic beam splitter and sent along separate paths before being recombined in the time-of-flight (TOF) spectrometer. The blue energy was controlled simply by varying the incident energy on the KDP crystal, and the IR energy was varied using a half-wave plate and polarizer in the IR arm. The IR arm contained a translation stage which could vary the path length in 10-μm steps allowing a time resolution of 33 fs.

The blue and IR were focused separately at $f/10$. We calculate that group velocity dispersion in the delay line increases both pulse lengths by no more than 5 fs in the interaction region. The laser pulses were focused down into a low-density ($\sim10^{14}$ cm^{-3}) beam of clusters and the energies of the ions produced in the interaction were determined from measurement of their flight time in a field-free drift tube, as detailed in Ref. [7].

The translation stage position corresponding to zero time delay between the blue and IR pulses was found from ionization rates in monatomic xenon. The integrated Xe^{2+} and Xe^{3+} ion yields were measured as a function of translation stage position. Numerical integration of the Ammosov, Delone, and Kräinov (ADK) tunneling ionization rates over the focal volume confirms that the ion yield peaks when the two pulses are coincident in time. By fitting a Gaussian to the

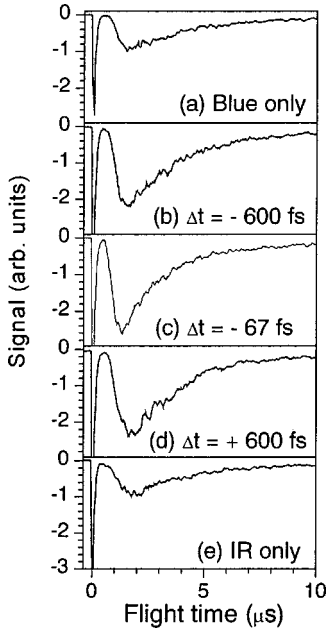

FIG. 2. Measured ion time-of-flight spectra from the irradiation of 14 000-atom Xe clusters with a 780- nm and/or 390- nm laser pulse. (a) Blue pulse only. (b) Blue precedes IR by 600 fs. (c) Blue precedes IR by 67 fs. (d) Blue pulse 600 fs after IR. (e) IR pulse only. In each case the intensity of the 780-nm pulse was 6×10^{14} W cm^{-2} pulse and that of the 390-nm pulse was 2.5×10^{15} W cm^{-2}.

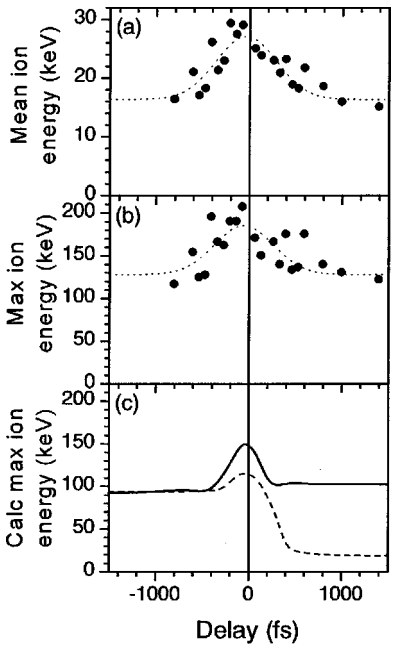

FIG. 3. (a) Mean and (b) maximum ion energies measured from the irradiation of 14 000-atom Xe clusters with one 780-nm, 6×10^{14} W cm^{-2} pulse (1.9 mJ) and one 390-nm, 2.5×10^{15} W cm^{-2} pulse (1.3 mJ) as the delay between the two pulses is varied. The energies obtained with the 780-nm pulse alone were 14-keV mean and 143-keV max. With the 390-nm pulse alone, the mean was 19 keV and 140 keV the maximum. The dotted lines are Gaussian fits to the data. (a) The center of the fit is at -40 ± 35 fs and the full width at half maximum (FWHM) is 765 ± 130 fs, (b) center at -60 ± 60 fs, FWHM 750 ± 200 fs. (c) Calculated ion energies. Clusters of 14 000 Xe irradiated by a 390-nm, 185-fs pulse at 2.5×10^{15} W cm^{-2} and a 780-nm, 260-fs pulse at either 4×10^{15} W cm^{-2} (solid line) or 6×10^{14} W cm^{-2} (dashed line).

data points, we were able to find the position corresponding to zero delay ($\tau=0$) to $\pm$ 10 fs. We define negative delay times to mean the blue pulse precedes the IR pulse.

We have obtained ion time-of-flight spectra from the explosion of ~14 000-atom (6-nm radius) Xe clusters irradiated sequentially by 6×10^{14} W cm^{-2}(1.9 mJ) at 780 nm and 2.5×10^{15} W cm^{-2}(1.3 mJ) at 390 nm as the delay between the two pulses was varied (Fig. 2). The relative intensities of the blue and IR pulses were set to give approximately the same mean ion energy individually. The observed time-of-flight spectrum changes with the relative delay between the two laser pulses. At $\tau=-67$ fs [Fig. 2(c)], the peak of the spectrum shifts to earlier arrival times, corresponding to higher kinetic energies.

The mean and maximum ion energies calculated from the time-of-flight spectra (as described in Ref. [7]) are shown in Figs. 3(a) and 3(b). Both the mean and the maximum ion energy vary with the delay between the two pulses for delays less than ~1 ps. A Gaussian fit to the data points puts the optimum delay at $\tau=-40\pm35$ fs or $\tau=-60\pm65$ fs (blue pulse precedes the IR) from the mean and maximum ion energies, respectively. The mean and maximum ion energies at the optimum delay are 29 and 210 keV, respectively, approximately double those obtained with either pulse alone.

With a single 780-nm laser pulse, the measured ion energies increase rapidly with incident laser intensity for intensities up to $\sim6\times10^{14}$ W cm^{-2}, and then vary as $\sim I^{0.5}$ at higher laser intensities. A laser energy of 5.9 ± 0.3 mJ at 780 nm would be required to produce ions with a mean ion energy of 29 keV, nearly twice the total energy of 3.2 mJ (1.9 mJ IR+1.3 mJ blue) in the two-color case. The pulse energy at 390 nm required to produce ions of this energy is well beyond the maximum we were able to attain. However, we expect a similar intensity scaling at both wavelengths and estimate that 4 mJ of 390-nm radiation would be needed. This confirms that a correctly timed sequence of two pulses of different frequencies can indeed be used to enhance the cluster ion energies.

If we assume that the optimum delay is when the period of resonant heating is at the peak of the laser pulse, we can estimate the temporal variation of the electron density at that point in the cluster expansion. The electron density drops from $3n_{\rm crit}$ at 390-nm (2.2×10^{22} cm^{-3}) to $3n_{\rm crit}$ at 780 nm (5.5×10^{21} cm^{-3}) in ~50 fs, a drop of 3.3×10^{20} cm^{-3} fs^{-1}. This drop is due to a combination of electron free-streaming and cluster expansion.

Figure 3(c) shows the results of the numerical model for the case of a 14 000-atom Xe cluster irradiated by a 390-nm pulse at an intensity of 2.5×10^{15} W cm^{-2} and a 780-nm pulse at 4×10^{15} W cm^{-2} (solid line) or 6×10^{14} W cm^{-2} (dashed line). The calculated optimum delay is at $\tau=-40$ or -55 fs for the two cases, respectively, consistent with the experimental results. The calculated ion energies agree reasonably well with the measurements, given the factor of ~3

uncertainty in the intensity calibration.

We have compared the ion energies produced in the cluster explosion when the two laser fields have parallel and perpendicular polarization. The measured mean ion energies as a function of delay between the two laser pulses were found to be the same (to within the error on the mean ion energy) whether the blue and IR pulses had parallel or perpendicular polarizations. This implies that the explosion is not driven by a process that is strongly dependent on the peak value of the electric field. This is consistent with the nanoplasma model, as the heating mechanism (collisional ionization) is not strongly dependent on the peak electric field strength. The tunneling ionization rate is, however, strongly dependent on the peak electric field strength. In the nanoplasma model, tunneling ionization is only important at the start of the explosion. It provides a seed for subsequent thermal collisional ionization, but the final ion energies are not strongly dependent on the numbers of tunnel ionized electrons. In contrast to this, the ionization ignition model [9] rests on the enhancement of the tunnel ionization rate due to the close proximity of other charged ions in the cluster. In this case, the tunnel ionization rate would be significantly enhanced when the blue and IR fields have parallel polarizations compared to the case when they have orthogonal polarizations, in contrast to our measurements.

In conclusion, we have presented numerical simulations and experimental results which show a change in the cluster dynamics when the cluster is irradiated by two pulses of different frequencies in sequence. The cluster experiences two peaks in its heating and electron temperature, which leads to an increase in the energies of the ions produced in the explosion. In contrast, a cluster irradiated by a sequence of pulses of the same frequency will experience only one peak in the heating. We have measured the ion energies from Xe clusters irradiated at the laser fundamental frequency (780 nm) and its second harmonic. The mean and maximum ion energies are found to depend on the relative delay of the two pulses (being doubled for the optimum value of the delay) but not on their relative polarizations. The calculations successfully reproduce the trends in the measured ion energies as the delay is varied, predicting the degree of enhancement and the pulse sequence required to optimize the ion energies. Significantly less laser energy is required to obtain the same ion energies when the cluster is irradiated with two pulses of different frequencies rather than a single laser pulse. The increase in the ion energies which can be obtained through two-color irradiation may have applications which include boosting the yield of fusion neutrons, which have been observed in the explosion of laser-heated deuterium clusters [10].

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