Asymmetric High-Energy Ion Emission from Argon Clusters in Intense Laser Fields

V. Kumarappan, M. Krishnamurthy, and D. Mathur

Tata Institute of Fundamental Research, Homi Bhabha Road, Mumbai 400 005, India

(Received 26 February 2001; revised manuscript received 27 April 2001; published 6 August 2001)

Clusters of 2×10^3 to 4×10^4 Ar atoms are Coulomb exploded in intense (up to 8×10^{15} W cm^{-2}) laser fields. The dependence of multiply charged argon ion energies on the polarization state of light is probed. A directional asymmetry in the ion-explosion energies is observed for the highest charge states. The ion-energy distribution consists of a low-energy isotropic component, and a high-energy anisotropic one. The results are discussed in terms of an asymmetric Coulomb-explosion scenario.

DOI: 10.1103/PhysRevLett.87.085005 PACS numbers: 52.50.Jm, 52.38.Ph

Particle acceleration using tabletop lasers has been one of the driving forces behind intense field research [1]. Large clusters have been shown to form a unique target for high-energy particle generation, combining the advantages of gaseous (debrisless) and solid (high density) targets. Laser-cluster interactions have been shown to be extremely energetic due to the high local density and the lack of conductive dissipation of heat into a bulk substrate. Energy deposition rates of 240 mW per atom have been measured [2], leading to a violent breakup of the cluster, involving the emission of energetic particles and photons. The mechanism of absorption and dissipation of such large amounts of energy in the clusters is still not understood. Over the past few years, there has been considerable debate on various mechanisms that might result in the production of ions of energies up to 1 MeV [3] and electrons up to 3 keV [4] in laser-cluster interactions. This issue is closely related to that of the production of charge states much higher than those that the laser field alone would be able to produce if rare gas atoms were independently subjected to it. The various models proposed include the coherent electron motion model [5], the microplasma model [6], and the ionization ignition model [7]. While the plasma model is expected to be applicable to sufficiently large clusters, there is disagreement on what exactly constitutes a sufficiently large cluster. Ditmire *et al.* [6,8] have claimed, using both theoretical arguments and experimental data, that clusters larger than a few hundred atoms undergo hydrodynamic expansion, while Lezius *et al.* [9] claim a Coulomb explosion mechanism for argon clusters as large as 1.8×10^5 atoms.

We have recently reported the polarization state dependence of K-shell x-ray emission from argon clusters, which was found to be independent of the ellipticity of the light field [10]. In this Letter, we report the angle dependence of ion emission produced by the explosion of argon clusters of 2×10^3 to 4×10^4 atoms. While a small dependence of ion energies on the direction of the polarization vector has been reported [11], it was attributed to an asymmetric low-energy component of the electron spectrum. We show that our measurements are not consistent with such an interpretation. The ion energies are measured using a time-of-flight (TOF) technique for laser polarization both along the TOF axis and perpendicular to it. While a major part of the ion-energy spectrum is unaffected by the change in polarization, the highest energy ions are seen to depend on the direction of the electric field of the laser. We conclude from this data that there are two components in the ion-energy spectrum—a low-energy isotropic component and a high-energy anisotropic one. Existing models need to be revised to account for this asymmetry, and for the existence of two components. We propose a mechanism that involves the direct acceleration of the ions by the laser field, as well as asymmetric production of highly charged ions.

In our experiment, clusters are produced by a solenoid-driven nozzle, which can be backed with up to 14 bars stagnation pressure. The nozzle has an aperture of 500 μm and an opening angle of 30°. The cluster beam is collimated by a skimmer and transferred to a region of high vacuum where the pressure is maintained in the 10^{-7} Torr range. The duration of the pulse can be varied from 100 μs to 1 ms, and its repetition rate can be continuously changed from 0 to 50 Hz. The laser is a chirped-pulse-amplified titanium:sapphire system that operates at 806 nm, with 10 Hz repetition rate, 100 fs pulse duration, and delivers up to 55 mJ of energy. In this series of experiments, up to 10 mJ pulses were focused to intensities up to 8×10^{15} W cm^{-2} using a 25 cm planoconvex lens. The intensity calibration was verified by measuring the appearance threshold of Ar^{5+} ions produced from argon atoms. A half-wave plate placed just before the lens is used to control the polarization vector of the laser light.

The production of large clusters from the nozzle was verified by measuring the Rayleigh scattered signal from the clusters using the third harmonic (355 nm) of a picosecond Nd:YAG laser. Hagena's scaling law [12] was then used to estimate the size of clusters produced from our nozzle. The methodology has been described elsewhere recently [10]. We estimate the average size of clusters in these experiments to range from 2×10^3 atoms per cluster at a stagnation pressure of 2 bars to 4×10^4 atoms per cluster at 10 bars, which was the maximum used in this series of experiments. The laser beam intercepts the cluster beam 26 cm downstream of the skimmer. We

estimate the average number density of argon atoms to be about 10^{16} cm^{-3}, most of which is condensed to a density of 4.4×10^{21} cm^{-3} in the cluster beam. The ions that are formed upon irradiation of the clusters with the focused laser beam were detected by a channeltron at the end of a 58 cm long flight tube, which is perpendicular to both the laser and the cluster beams. The ion-energy spectrum is typically averaged over 500 laser shots. The high-energy ions are detected only when the laser and the gas pulse are temporally coincident. In front of the channeltron, three high transmission grids are used, with the middle grid charged to a positive voltage and the other two kept grounded, to analyze the charge states present in the ion-energy spectrum. The intergrid separation is 5 mm; the total length of the repeller assembly (10 mm) is small compared to the 580 mm flight tube. The channeltron cone is kept at -2500 V, so that electrons with energies lower than 2.5 keV do not strike the detector.

Figure 1(a) shows a typical ion-energy distribution from $Ar_{40\,000}$ clusters (10 bars) when irradiated with 8×10^{15} W cm^{-2} laser pulses. The energy distribution is calculated from the time-of-flight spectrum using the relation $f(E) = f(t)\,(dE/dt)^{-1}$. The highest energy seen is about 300 keV, and the mean energy, $\bar{E} = [\int Ef(E)\,dE]/[\int f(E)\,dE]$, of the ions is 13.2 keV. The highest ion energy is determined from the minimum in the raw TOF spectrum between the ion and the electron/photon feature. Note that there is a low-energy cutoff due to the finite record length of the digital oscilloscope; and, hence, the average does not include ions with energies lower than 180 eV. The general features of the spectrum are in good agreement with spectra reported earlier [13], where the experiment was similar in design. Curves 1(b) and 1(c) in the figure show spectra obtained with 200 and 400 V, respectively, on the repeller plate. When a voltage V is applied on the repeller plate, all ions with charge q and energy less than qV are unable to reach the detector. The TOF of ions with energies greater than this cutoff remains unaffected because the ions spend only a small fraction of their flight time in the repeller assembly. Therefore the spectrum shows a series of steps corresponding to the values of the charge state q. The height of the step indicates the yield of the charge q at energy qV. Curve 1(b) shows steps for Ar^{+} to Ar^{7+}, while in curve 1(c) Ar^{4+} to Ar^{8+} can be distinguished. Figure 2 shows a 2D spectrum constructed from a series of spectra taken with different repeller voltages. A two-dimensional matrix of TOF spectra was first acquired by ramping the repeller voltage using a digital-to-analog converter. The intensity distribution in time was transformed to a distribution in energy, and the 2D energy distribution was then differentiated with respect to the y axis to obtain this spectrum. Note that the y axis also represents the energy per charge on the ion. Hence, the various charge states appear as lines with slopes equal to $1/q$. In such a graph, a linear dependence of the energy on q would cause the mean energies of all the charge states to appear on a horizontal line. Analysis indicates that the dependence is approximately quadratic. This is in agreement with the ion-energy distribution measured by Lezius *et al.* [9] for somewhat larger clusters, where the authors had concluded that the clusters undergo Coulomb explosion. We could identify multiply charged ions up to Ar^{8+}, beyond which the spectrum could not be resolved into constituent charge states. While Lezius *et al.* had

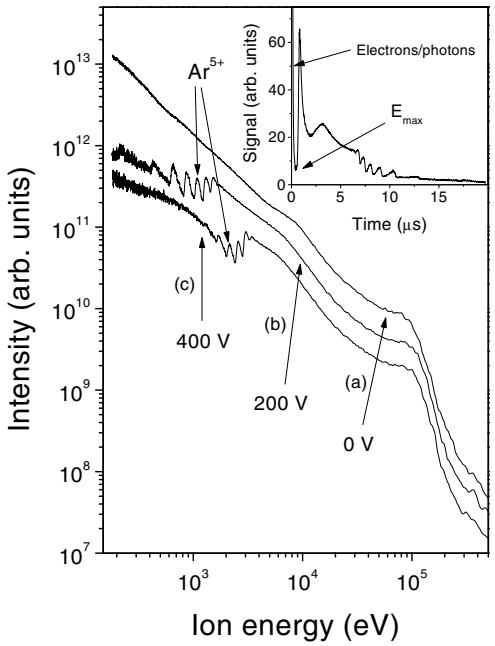

FIG. 1. Ion-energy distributions from $Ar_{40\,000}$ obtained with different repeller voltages. The distribution functions are vertically offset for clarity. Inset shows a raw TOF spectrum (200 V on the repeller). E_{max} denotes the highest ion energy detected. The laser intensity was 8×10^{15} W cm^{-2}, with polarization parallel to the TOF axis.

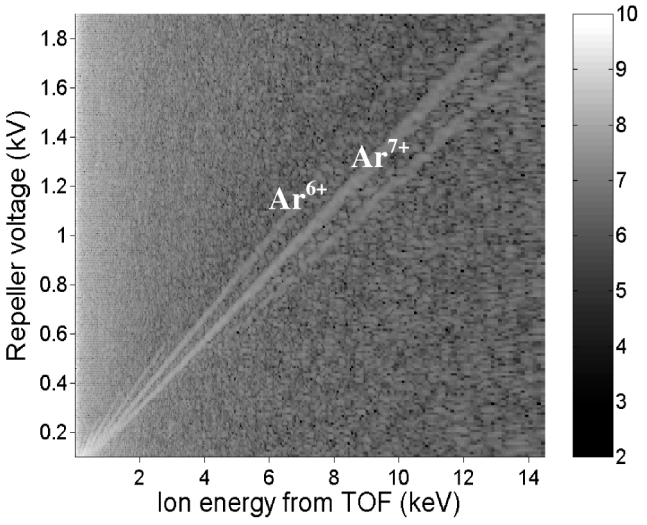

FIG. 2. Charge state resolved ion-energy distribution. The gray scale represents the logarithm of the distribution function. Each charge state q is seen as a line with slope $1/q$.

obtained better charge state resolution by using a magnetic deflection TOF spectrometer, they also could identify only up to Ar^{8+}. This is probably due to the significant difference in the ionization energies of Ar^{7+} (143 eV) and Ar^{8+} (422 eV).

Figure 3 shows spectra obtained when the polarization vector was rotated from a direction parallel to the TOF axis to one perpendicular to it. While a large part of the spectrum remained unchanged, there is a clear shift to higher energies at the highest end of the range. The ion yields for parallel polarization (0°) in the latter region were found to be larger than for perpendicular polarization (90°) by a factor of up to 2. It is important to note that the ion spectrum does not shift uniformly to higher energies, only at the highest energies is a shift evident. This was verified by measuring the energies with a voltage applied to the repeller plate. Figure 3 shows spectra obtained at 100 V for both polarization directions. In these spectra, the height of the step at the cutoff energy indicates the ion yield at that energy. Clearly there is no difference in the ion distribution at low energies between the two polarizations. Measurements were made over a range of cluster sizes (5000–40 000 atoms per cluster) and intensities (10^{15}–10^{16} W cm^{-2}) and similar behavior was observed over these ranges. The laser intensity was monitored on a shot-to-shot basis for these measurements, and it was ensured that the fluctuations were less than 5%.

It follows, from the dependence of the ion energies on the charge state and the absence of asymmetry in low-energy ion emission, that only the highly charged states are emitted nonspherically. Shao *et al.* [4] have measured the angular dependence of electron emission from xenon clusters, and they observe an anisotropic "warm" component and an isotropic "hot" component. Springate *et al.* [11] have also reported an anisotropy in the ion emission, which they attribute to the influence of the warm electrons. If, following [11], we assume that the asymmetry we observe is due to the low-energy component of the electron spectrum, we run into two contradictions. In the hydrodynamic expansion model, it is the hydrodynamic pressure of the laser-heated electrons that drives the explosion of the cluster [6]. But, as shown in [9] and indicated in Fig. 2, argon clusters in this size range explode due to Coulomb forces, and the electron angular distribution should, therefore, not influence the ion angular distribution significantly. Moreover, an asymmetric electron distribution in the plasma model should lead to asymmetric distributions for *all* charge states, a conclusion not supported by experiments for the clusters studied here. In particular, the *low-energy* electrons cannot cause an asymmetry only in the *high-energy* ion distribution.

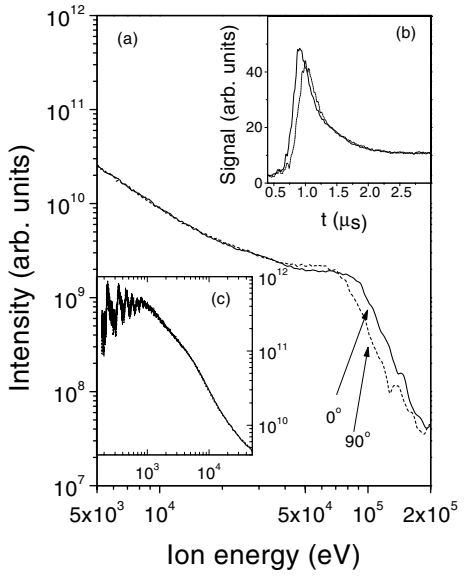

FIG. 3. (a) Ion-energy spectra from $Ar_{40\,000}$ for laser polarizations parallel (0°) and perpendicular (90°) to the TOF axis. Note that, for energies less than 4×10^4 eV, the two spectra overlap; differences are seen only for high-energy components. Inset (b) shows corresponding raw TOF spectra. Inset (c) shows spectra obtained after cutting off part of the low-energy component (100 V on the repeller).

Recently, Ishikawa and Blenski [14] have claimed that the charge state of an ion may change rapidly in resonance with the optical field. The radial field due to the charged cluster and the laser field act in the same direction for half the laser cycle, and a high charge state can be reached. But in the next half cycle, the two fields oppose each other, and this charge state can no longer be maintained. Recombination in this period leads to a lower charge state, and, hence, a smaller force on the ion due to the laser field. The cycle-averaged force on the ion due to the laser field itself could, in such a situation, be significant, and the laser itself could accelerate the ions along the polarization vector. We have computed that, for a 100 fs rectangular pulse at an intensity of 2×10^{16} W cm^{-2}, this force alone can accelerate an argon ion to $q \times 30$ keV, where q is the average difference in the charge states during two half cycles of the laser pulse. Thus, this mechanism could produce an anisotropy of the same order of magnitude as has been observed. But, contrary to our measurements, the model also seems to predict that all charge states should be anisotropically accelerated.

Kou *et al.* [15] have proposed a model (for C_{60}) in which the laser has no role in the acceleration of the ions, but the charge state distribution produced by the laser is asymmetric, and, hence, so is the ion-energy distribution. According to this model, electrons "cascade" through the nuclear potential wells under the influence of the laser field. Molecular dynamics simulations show that this process leaves the polar regions (the axis being defined by the laser field) with higher charge states. Subsequent Coulomb explosion leads to an anisotropy in the ion spectrum. While our observations are consistent with the expectations of this model for the highest charge states, the model also predicts that lower charge states should be emitted preferentially

along the direction perpendicular to the laser field. Such an anisotropy has not been observed in our experiments. We note that, in the case of C_{60}, C^{2+} is expected to be emitted perpendicular to the laser polarization direction because it is depleted at the poles and, hence, is predominantly emitted from the equator. For a cluster much larger in size, such depletion might not be large enough to be measurable in an experiment of this kind.

Although these two models seem to be quite distinct, we note that they are not mutually exclusive. Indeed, Figs. 8(d) and 8(e) of Ref. [15] show a clear change in the charge states at the poles in half a cycle of the laser field. Both effects act together to produce an anisotropy in the ion emission. We present in Fig. 4 a schematic view of the cluster for two consecutive half cycles. In the first half cycle, the bottom pole is depleted of electrons due to the combined action of the laser field and the radial electric field. In the second half cycle, the electrons are driven back towards the bottom pole, leaving the pole at the top highly charged. This alternation of charge states results in a net cycle averaged force on the ions at the poles. Moreover, on the average, the charge state at the poles is higher than in the rest of the cluster. This leads to an asymmetric Coulomb explosion force, driving the high charge states preferentially along the direction of the laser polarization. The ions near the surface of the cluster would be most strongly influenced by this competition between the laser field and the radial field, where the latter peaks, and the former is not reduced by shielding due to electrons. The ions originating in the bulk of the cluster would, hence, show less anisotropy. The difference between the surface ions, particularly those at the poles of the cluster, and the bulk ions manifests itself in the distinct angular distributions for these two groups of ions. All our experimental observations can be rationalized using this scenario.

In conclusion, we have found that, following a cluster explosion, the ion-energy spectrum has two components—a high-energy one that depends on the laser polarization, and a low-energy one that is independent of it. The high-energy component exhibits asymmetry, and the ion yield is greater in the direction of the laser field. Our observations can be explained by an asymmetric Coulomb explosion, aided by the direct acceleration of the ions by the laser field; tantalizing possibilities open of forming pulsed beams of energetic ions.

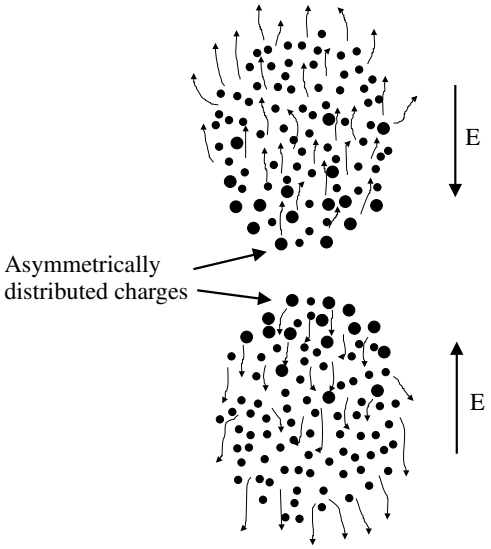

FIG. 4. Schematic representation of the distribution of ion charge states on a cluster during two consecutive laser half cycles. The large dots represent the asymmetrically distributed highly charged states. The arrows represent electron trajectories.

We thank G. Ravindra Kumar for useful discussions. Support from the Department of Science and Technology for our laser system is acknowledged.

[1] See, for instance, M. I. K. Santala *et al.* Phys. Rev. Lett. **86**, 1227 (2001), and references therein.
[2] K. Boyer *et al.,* J. Phys. B **27**, 4373 (1994).
[3] T. Ditmire *et al.,* Nature (London) **386**, 54 (1997).
[4] Y. L. Shao *et al.,* Phys. Rev. Lett. **77**, 3343 (1996).
[5] B. D. Thompson *et al.,* J. Phys. B **27**, 4391 (1994).
[6] T. Ditmire *et al.,* Phys. Rev. A **53**, 3379 (1996).
[7] C. Rose-Petruck *et al.,* Phys. Rev. A **55**, 1182 (1997).
[8] T. Ditmire *et al.,* Phys. Rev. Lett. **78**, 2732 (1997).
[9] M. Lezius *et al.,* Phys. Rev. Lett. **80**, 261 (1998).
[10] V. Kumarappan, M. Krishnamurthy, D. Mathur, and L. C. Tribedi, Phys. Rev. A **63**, 023203 (2001).
[11] E. Springate *et al.,* Phys. Rev. A **61**, 063201 (2000).
[12] O. F. Hagena and W. Obert, J. Chem. Phys. **56**, 1793 (1972).
[13] T. Ditmire *et al.,* Phys. Rev. A **57**, 369 (1998).
[14] K. Ishikawa and T. Blenski, Phys. Rev. A **62**, 063204 (2000).
[15] J. Kou *et al.,* J. Chem. Phys. **112**, 5012 (2000).