Explosion Dynamics of Rare Gas Clusters in Strong Laser Fields

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We have studied the ionic outcome from the interaction of intense laser light with large argon and xenon clusters. Ions with initial energies of several 100 keV are charge and energy selected using a magnetic deflection time-of-flight mass spectrometer. For argon clusters, Coulomb repulsion is the key process in the explosion mechanism, whereas for xenon we observe a mixture of Coulomb repulsion and hydrodynamic expansion. Coulomb explosion is the preferred decay channel for smaller clusters and it is also responsible for the production of the most energetic ions. Our results can be understood on the basis of a charged sphere model of cluster-sized plasmas. [S0031-9007(97)05135-1]

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The development of relatively compact but powerful short pulse lasers has given strong impetus to the study of matter in intense fields. Solid and gaseous targets have been thoroughly investigated and, generally, highly excited states of matter are reported (see, for example, [1]). Recently, since atomic clusters have an intermediate target size, they have captured the attention as a promising means to produce x rays in the keV range [2–4], high particle velocities [5,6] and highly charged ions [6–8]. According to Wülker *et al.* [9] as well as Ditmire and co-workers [10,11], the very energetic states of matter obtained when irradiating clusters are usually interpreted as hot cluster-sized plasmas, efficiently laser heated by inverse bremsstrahlung. Already for intensities of 10^{14} W/cm^2 we have been able to confirm hydrodynamically expanding nanoplasmas with ion energies of several keV [8]. Much higher particle energies reaching up to 1 MeV, obtained after irradiating xenon clusters with 10^{16} W/cm^2, are also currently attributed to hydrodynamic expansion [5,6]. It should be mentioned that, after the pioneering work by Ehler [12], ion energies of several MeV/amu have been reported from intense laser irradiation of solids for more than a decade [13,14]. Such energetic particles have been theoretically interpreted as a departure from ideal isothermal expansion due to overheated coronal electrons [15], rarefaction shocks [16], neutralizing return currents [17], ion acoustic turbulences [18], or self-focusing [19]. Most of the theoretical studies on fast ion generation have assumed that the energy distribution in the coronal plasma is described primarily by two temperatures for thermal and hot electrons. Interestingly, two electron temperatures have also been observed from clusters, although electron energies generally did not exceed 3 keV [20]. A difference in three orders of magnitude between ion and electron energies obtained from similar cluster experiments [5,20] could therefore again indicate a failure of the isothermal expansion mechanism. From previous studies on molecules [21] and small clusters [22], a transition regime can be expected for shrinking particle sizes towards a Coulomb explosion mechanism originating from the charge buildup in the system due to electron losses. In such a case, the derivation of initial electron and ion temperatures from their respective final energies will be affected, which may have consequences for future experiments and applications. To decide when the dynamic of cluster-sized plasmas is governed by hydrodynamic and when by Coulomb mechanisms, it is necessary to obtain detailed maps of the charge dependence of the ion energies. In this Letter, we study the interaction of intense laser light with argon and xenon clusters using magnetic deflection time-of-flight (MD-TOF) mass spectrometry. With this technique, highly energetic (up to 1 MeV), multiply (30+) charged ions can be successfully analyzed. We demonstrate that in the case of argon the explosion dynamics appears to be purely governed by Coulomb repulsion. Moreover, in the case of large xenon clusters we obtain fragment energies resulting from both Coulomb explosion and hydrodynamic expansion. Our data suggest that very energetic highly charged ions arise from ejection near the surface of a cluster-sized precharged plasma. It is interesting to note that highly energetic ions produced from laser-solid interactions could also result from a similar mechanism since laser ablation usually leads to intense cluster production [23].

The basic experimental setup has been described in detail very recently [8]. Large clusters are produced by supersonic expansion of the gas through a 150 μm conic nozzle. As this source is identical to that used in elaborate cluster size measurements, we can estimate average sizes $\bar{N}$ to be in the range of 1.8×10^5 for argon (16.5 bars) and 2.0×10^6 for xenon (15.5 bars) [4,24]. Thus, most of the matter in the supersonic beam with an average of 10^{14} atom/cm^3 is concentrated into local densities between 3.5×10^{21} cm^{-3} (Xe) and 4.4×10^{21} cm^{-3} (Ar). The beam is irradiated with Ti-sapphire laser pulses ($\tau = 130$ fs, $E_L = 60$ mJ, $\lambda = 790$ nm) which are focused with a $f = 170$ mm off-axis parabolic mirror into the center of the first acceleration region of a double chamber Wiley-McLaren TOF [25]. From optical field ionization of neon we estimate the maximum laser intensity to $5 \times 10^{17} \pm 20\%\ \mathrm{W\,cm^{-2}}$ [26]. The TOF extraction fields F_1 and F_2 are 375 and 2250 V/cm, respectively. Thus, ion startup energies E can usually be obtained from the symmetric peak splitting $\pm\Delta t/2$ of energetic forward and backward ions using $E = (qF_1\Delta t)^2/m$ [25,27]. However, in the case of startup energies up to several 100 keV and charge states up to 30, the mass resolution of a double chamber TOF is usually not sufficient. Additionally, it is not always possible to measure Δt because reflected ions can also be absorbed by the repeller plate [8]. Besides, we observe that recombination of highly charged ions on the chamber walls generates UV photons that can severely contaminate ion detection. Consequently, we have introduced a grounded slit (1×5 mm) in the center of the TOF tube, such that only ions passing along the axis defined by the laser focus and the metal slit are detected. Behind the metal slit a weak homogeneous magnetic field ($B = 2630 \pm 50$ G, length 35 mm) has been introduced. The exterior of the magnetic region is efficiently screened with iron and the deflection unit can be positioned between 20...120 mm relative to the detector. For ion detection, the high sensitivity electron multiplier is mounted on a precision translator and shielded entirely with a metal case, except for a 1×5 mm ion entrance slit. In comparison to a recently presented single shot magnetic deflection spectrometer [28], our simple and inexpensive solution is well adapted for integration over numerous laser shots, thereby leading to an enhanced signal-to-noise ratio.

If the detector is positioned at a distance l from the entrance into the magnetic field of length d, for small deflection angles α, an ion with charge q, mass m, and total kinetic energy E_{tot} will be deflected by

$$x = Bd(l - d/2)\sqrt{q^2/(2mE_{\mathrm{tot}})}\,. \tag{1}$$

Thus, from the measurement of x the startup energy E can be obtained directly from the total electrostatic potential in the focal volume ($U = 3375$ V) using $E = E_{\mathrm{tot}} - qU$. On the other hand, the TOF t of forward and backward ions with energy E can be calculated according to Wiley and McLaren [25]. For forward ions with large startup energies, qt/x reaches a constant value. For large deflection angles, Eq. (1) becomes invalid, and numerical integration of the equations of motion of the ions must be performed. Thus, to analyze our experimental data we have simulated ion flight times and deflection angles using time steps of 5 ps with an energy conservation better than 99.9%.

Figure 1 shows an experimental MD-TOF spectrum obtained after irradiation of argon clusters. Forward and backward scattered ions with $q = 1\ldots8$ are fully resolved. The startup energies of these ions are evaluated directly from the peak splitting and can be modeled with $E(q)[\mathrm{eV}] = q^2\alpha_c$, assuming $\alpha_c = 160$ eV, thus indicating a Coulomb explosion mechanism. For comparison, simulated x and t of ions with such an energy dependence are included in the figure. Moreover, it is interesting to note that weak ion signals marked with A in Fig. 1 are produced by highly energetic ions with $q > 6$ and startup energies up to 50 keV. It is clear that the ion energies cannot be understood with a hydrodynamic expansion model, where $E(q) = qkT_e$ is determined by the electron temperature. We conclude that, to reach the hydrodynamic regime, it is necessary to increase particle size and electron temperature. Electron heating by intense light is expected to rise proportional to Z^2 for increasing atomic number Z of the target constituents [29]. Moreover, as ion heating basically originates from electron-ion collisions after excitation of the electron plasma, collisional frequency and thus heating efficiency should rise proportional to $\bar{N}^2$ [4]. Following such considerations, we have studied xenon clusters that allow us to obtain about ten times larger particle sizes compared to the argon case while leaving the overall atom density in the focus intact. The latter is a necessary condition to avoid secondary space charge effects, thus making it useless to increase simply the argon cluster size by using higher stagnant pressures. Figure 2 shows a MD-TOF spectrum of xenon. Ions with $q < 3$ again show forward-backward split signals, and their energies can be analyzed directly from Δt. For higher charge states, reflected signals are lost at the repeller. Most interestingly, analysis of the ion energies indicates that two distinct acceleration mechanisms are involved. Ions with $q < 6$ seem to follow a nonlinear Coulomb driven charge dependence with $E(q)[\mathrm{eV}] = q^2\alpha_c$, with $\alpha_c = 180$ eV. In contrast, most of the ions with $q > 10$ produce a hydrodynamic extraction energy pattern with $T_e = 1.5$ keV. The highest detectable charge state is evaluated at 30+ by our simulation. Furthermore, ion signals of

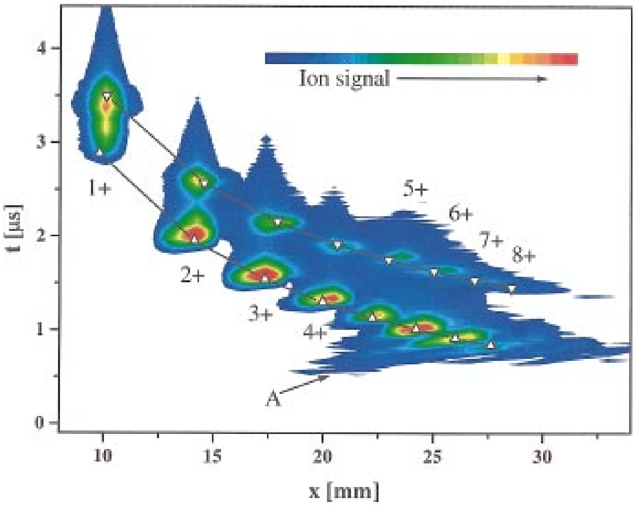

FIG. 1(color). Two-dimensional contour plot, showing a MD-TOF spectrum obtained from laser irradiation of large argon clusters. Signals correspond to Ar^{q+} ions, ($q = 1\ldots8$). The startup energies E for forward ($\triangle$) and backward (∇) ions are simulated numerically using $E(q) = 160[\mathrm{eV}]q^2$.

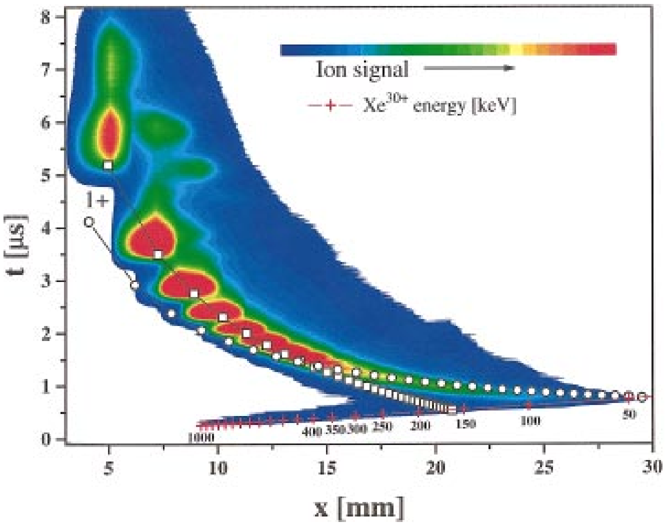

FIG. 2(color). MD-TOF spectrum obtained from laser irradiation of large xenon clusters. Signals correspond to forward Xe^{q+} ions, ($q = 1 \dots 30$), whereas weak backward signals are only seen for $q = 1, 2, 3$. Ion energies are simulated using a hydrodynamic approach {○: $E(q) = \alpha_h q, \alpha_h \approx 1.5$[keV]} or the charged sphere model {□: $E(q) = \alpha_c q^2, \alpha_c \approx 180$[eV]}. The inserted energy scale ($- + -$) indicates the position of Xe^{30+} ions with startup energies between 50 keV and 1 MeV.

charge states between 20+ and 30+ show an ultrahigh energy tail that is only weakly deflected by the magnetic field. For better comparison, we have included an energy scale calculated for Xe^{30+} ions, and it can be seen that such signals apparently arrive at x-t positions, indicating startup energies reaching 1 MeV. Whereas such startup energies present the upper limit of our present observations, the lower limit is determined to be ≈400 keV, assuming Xe^{25+}. Note that such fast ions cannot be understood by hydrodynamic expansion, as for $q < 20$ no similar tails are observed. Instead, such high energies coincide well with the Coulomb explosion mechanism, an observation which is in contrast to previous interpretations in Refs. [5,6].

Furthermore, it can be seen in Fig. 2 that the simulated x reproduces the experiment very well, but especially for $q < 3$ the arrival time t is up to 800 ns delayed if compared to the ion energies. We observe that such a retarding effect depends on the backing pressure, and we suggest that for average cluster sizes exceeding 10^6 the plasma polarizability in the focal zone is able to shield extraction fields efficiently for several hundred nanoseconds. Such effects, which will be treated elsewhere, can, in principle, be understood when applying the polarizability derived from a Drude model to an expanding initially dense plasma. However, it is interesting to note that low charged ions are particularly sensitive to plasma retarding because they leave the focal zone with the smallest velocity. In contrast, it appears that highly charged particles are expelled so rapidly that the delay becomes negligible and determination of E and q from the x-t positions is valid for $q > 6$.

A particular advantage of MD-TOF spectrometry is that energy distributions for a given charge state can be directly obtained by parametrization of $t(E, q)$ and $x(E, q)$. For a set of energies between 1 eV and 1 MeV we have simulated t and x for charge states between 1 and 30, and measured the signal intensities along these points. Such energy distributions derived from the MD-TOF spectrum (Fig. 2) are depicted in Fig. 3. Charge states with $q > 12$ show the Coulomb driven high energy tail, which extends up to several 100 keV. We suggest that the substantial energy broadening results from local variations of the charge density per cluster.

Comparable Coulomb explosion mechanisms have long been observed in the strong field physics of small molecules. However, usually ion energies below 100 eV have been reported [21]. Such studies are generally performed using low density targets to explore the single molecule response. In contrast, collective phenomena that are expected to occur within clusters lead to a more complicated situation. Very recently, Brewczyk *et al.* [30] have applied a Fermi liquid model to small 1D Xe clusters in a strong electromagnetic field. They predict shell-by-shell ion ejections with very high velocities which basically originate from the initial Coulomb energy. The process is found to occur within a few optical cycles and indicates that, in addition to the loss of free streaming electrons, local Coulomb energy may arise from a deformation of the electron cloud with respect to the cluster core.

In addition, it is possible to describe Coulomb explosion using a simple charged sphere approach. In this model the irradiated cluster is considered as a perfectly conducting sphere with radius r. Then, the loss of electrons will lead to a charge buildup Q on the cluster surface that can be associated with the Coulomb pressure on the

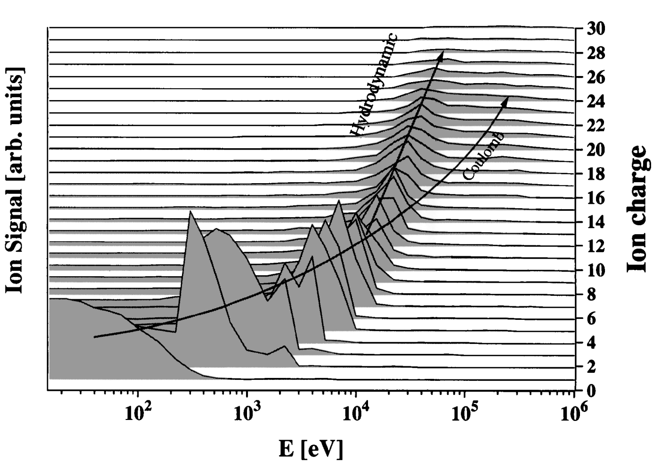

FIG. 3. Startup energy distributions of different xenon charge states ($q = 1 \dots 30$) deduced from the MD-TOF spectrum shown in Fig. 2 (see text).

sphere:

$$P_C = \frac{3Q^2}{2(4\pi)^2 \epsilon_0 r^4}. \tag{2}$$

It is possible to compare the Coulomb pressure to the hydrodynamic pressure $P_H = n_e(r) k T_e(r)$. Suppose that the radial velocity of the expanding plasma with a density $n_e(r) \propto r^{-3}$ reaches a constant value shortly after the laser pulse, $T_e(r) \propto r^{-1}$ solves the hydrodynamic differential equation [11],

$$\frac{\partial T_e}{\partial t} = \frac{-2T_e}{r} \frac{\partial r}{\partial t}. \tag{3}$$

This implies that P_C and P_H will be competing processes over a long expansion time since they both decrease with $1/r^4$. Moreover, precharging should depend primarily on the energy of the hot electrons in the plasma corona which can be estimated according to Ref. [29] to be in the range of 20...100 keV. Only electrons with energies E_e higher than the Coulomb potential are able to leave the system with radius r and, consequently, the maximum charge density on the sphere n_q can be estimated using

$$n_q = \frac{3\epsilon_0 E_e}{e r^2}. \tag{4}$$

Thus, Coulomb pressure will be superior in smaller clusters. This compares well to our present observation that the Coulomb energies are similar for very different cluster sizes (160 and 180 eV) but that the overall expansion behavior changes dramatically. We finally suggest that during the initial stage of the expansion the highest charged ions are quickly driven to the outer layer of the plasma as they are most efficiently accelerated. As soon as they reach the plasma vacuum edge they are rapidly ejected. Regarding the experimentally obtained values of α_C, we estimate a local Coulomb pressure per Van der Waals sphere of $\approx 10^{11}$[Jm^3], which corresponds to an average charge state between 13 for argon and 15 for xenon derived from Eq. (2). If we take electron ion recombination during the expansion into account, such values correspond well to the mass spectrometric result [7] and recent x-ray spectroscopic data [4].

In summary, different explosion mechanisms are observed for laser irradiated rare gas clusters. Whereas argon clusters undergo pure Coulomb repulsion, xenon exhibits a mixed Coulomb-hydrodynamic expansion behavior. In particular, ion energies resulting from hydrodynamic expansion of large xenon clusters indicate electron temperatures in the range of 1.5 keV. However, the most energetic ions arise from Coulomb repulsion. Clearly, the observed quadratic charge dependence of the ion energies cannot be understood using common plasma corona theories [15–19]. Instead, we propose a simple Coulomb explosion model of a precharged cluster-sized plasma which is in good agreement with the present experimental results.

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