Anisotropic "charge-flipping" acceleration of highly charged ions from clusters in strong optical fields

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Measurement of energies of ions that result from disassembly of atomic and molecular clusters [Ar_n, $n=2000-40\,000$; $(N_2)_n$, $n=50-3000$] in strong optical fields provides evidence for "charge-flipping" acceleration that gives rise to ions with energies in excess of the Coulombic limit. Measurements of ion spectra as a function of cluster size, and as a function of laser polarization, demonstrate different facets of charge-flipping acceleration.

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I. INTRODUCTION

The physics that governs the disassembly of atomic clusters in intense optical fields has attracted considerable attention in the course of the last decade (for a recent review, see Ref. [1] and references therein). Large atomic clusters, consisting of several hundred to several hundred thousand atoms per cluster, provide the high density required for substantial and efficient absorption of laser energy. While the energy is absorbed primarily by the electrons in the cluster, it is rapidly redistributed in the form of incoherent radiation (few keV x rays) and highly energetic ions. While maximum ion energies as high as a MeV have been reported [2], mean energies are typically in the range of 10–100 keV. These are several orders of magnitude higher than the energies that are measured for small molecules. Existing theoretical models of cluster disassembly in strong optical fields succeed in only partially accounting for the experimentally observed features that presently drive research in this area. We report here on an experimental study of an ion acceleration mechanism that has not been *quantitatively* considered before. Atomic and molecular clusters of different sizes [Ar_n, $n=2000-40\,000$; $(N_2)_n$, $n=50-3000$] have been irradiated by strong optical fields and give rise to ions whose energies extend well beyond the Coulomb limit. We have made an experimental determination of the Coulomb limit. Ion energies are measured that extend beyond this limit, and it is this range of energies that have not been accounted for before and constitute the focus of attention in this work. We have also carried out an experimental test of the "charge-flipping" mechanism that is postulated to account for the energies beyond Coulomb limit. The consequences arising from these three hitherto-unaddressed facets are of general interest because of the intrinsic importance of studying the nonperturbative physics of light-matter interactions in situations far from equilibrium, and due to potential applications of fast, highly charged ions in fusion science, cancer therapy, and ion acceleration technology.

Prevailing understanding of laser-cluster interactions in the strong-field regime may be summarized in the following simple terms. Most atoms in a cluster are tunnel ionized at the leading edge of the incident laser pulse. Electrons that are ejected from individual atoms are accelerated away from the cluster by the ponderomotive potential and what gets left behind is a positively charged core that gives rise to an increasing potential barrier to further removal of electrons. The question of whether such a barrier is sufficient to retain a large fraction of electrons or not is a major point of contention between the two major models of laser-cluster interaction. In the hydrodynamic expansion model, it is assumed that retention of most of the electrons by the cluster results in a spherically symmetric, electrically neutral plasma. The retained electrons absorb energy from the laser by collisional inverse bremsstrahlung. The hot electron plasma expands due to hydrodynamic pressure, and transfers energy E to the ions, where $E=qkT_e$, T_e being the electron temperature. In the alternative scenario, the Coulomb explosion-ionization ignition model, the electrons that are ejected following tunnel ionization rapidly leave the vicinity of the cluster. As a result, there is a buildup of charge on the cluster that gives rise to a radial field that may become large enough to drive further ionization at the surface of the sphere. The removal of electrons at the surface increases the radial field further and "ignites" ionization. Cluster explosion due to the Coulombic repulsion between the positively charged ions then occurs and ion energies are given by $E \propto qQ/R$, where q is the total charge on the cluster of radius R and Q is the charge on the fragment ion formed upon cluster disassembly.

Neither of these scenarios accounts for several facets of the physics of cluster-laser interactions that have been unveiled in recent experiments. Among them are the following: (i) Asymmetric Coulomb explosion of clusters [3]; (ii) enhancement in the asymmetry of both the ion and electron emission at the resonance condition, where the plasma frequency matches the driving laser frequency and the energy absorption by the clusters is most effective [4]; and (iii) the temporal width of the resonance that is experimentally observed is much larger than that predicted by a uniform-density hydrodynamic model [5]. Recent results have provided the impetus for developing models that extend beyond the prevailing, somewhat simplistic, picture of isotropic, one-dimensional plasma expansion and of probing the possi-

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bility that new mechanisms for charged particle acceleration, over and above that due to (a) Coulombic fields in small and moderately sized clusters and (b) hydrodynamic pressure in large clusters, contribute to the overall dynamics when strong optical fields interact with clusters. We are not aware of any study that addresses the physics issues that determine the *maximum* energy with which ions are ejected following cluster disassembly.

We have applied time-of-flight (TOF) spectrometry to probe the disassembly of relatively small atomic and molecular clusters in strong optical fields. Our results provide experimental evidence for an acceleration mechanism that we designate charge-flipping acceleration. We confirm earlier results that the distribution of energies of ions ejected upon cluster disassembly has two components: a low-energy, isotropic component and a high-energy, anisotropic component. We show that the latter results from charge-flipping ion acceleration and results in ion energies that are *larger* than those expected from purely Coulombic considerations. Measurements of ion spectra for varying sizes of atomic and molecular clusters, and as a function of laser polarization, demonstrate different facets of charge-flipping acceleration.

II. EXPERIMENT

Our experiments were conducted using a supersonic jet expansion source that has been described in earlier reports [3,4,6]. Briefly, a piezodriven solenoid valve, with a 30° conical opening of 500 μm, was backed with stagnation pressure of up to 14 bars. The central part of the cluster beam was sampled by a 500 μm skimmer placed ~30 cm downstream, in an interaction chamber maintained at 10^{-7} Torr. A Ti:sapphire laser was used that produces 100 fs long pulses, at a repetition rate of 10 Hz, with a maximum energy of 55 mJ. For the present measurements, 10 mJ pulses were focused to intensities of 10^{16} W cm^{-2} by means of a 25 cm planoconvex lens. A half-wave plate placed just before the lens controlled the polarization vector of the laser light. Laser-cluster interactions occurred ~26 cm downstream of the skimmer. The ions formed when the laser and cluster beams were temporally coincident in the interaction zone were detected by a channel electron multiplier (CEM) placed at the end of a 58 cm long flight tube which was mutually orthogonal to the laser and cluster beams. Cluster formation was verified by measuring the Rayleigh scattered signal from the clusters using 355 nm light from a Nd:YAG (yttrium aluminum garnet) laser. Hagena's scaling law [7] was used to estimate cluster size; the average size of $(N_2)_n$ clusters ranged from $n=50$ at a stagnation pressure of 2 bars to $n=3000$ at 10 bars; Ar_n clusters ranged in size from $n=2000$ to $n=40\,000$ over the same range of stagnation pressure.

III. RESULTS AND DISCUSSION

Figure 1 shows the ion energy distribution for two $(N_2)_n$ cluster sizes at an intensity of 7×10^{15} W cm^{-2}, when the laser light is polarized along the TOF axis as well as perpendicular to it. There is a high-energy component in the distribution (at energies $\geqslant 3-5$ keV) that exhibits a distinct asymmetry: ion energies and fluxes are enhanced when the laser polarization vector is parallel to the TOF axis. The low-energy component of the distribution (at energies $\leqslant 3-5$ keV) is found to be isotropic. This type of behavior is consistent with that observed in atomic clusters [3,4]. However, with $(N_2)_n$ clusters, there is an additional feature: a very low-energy component (at energies $\leqslant 40$ eV) that is also asymmetric (see the inset in Fig. 1). This component has no analog in measurements with atomic clusters and we ascribe it to the ionization signal from unclustered N_2 molecules that are present in the supersonic beam. The asymmetry that is observed is attributed to spatial alignment of the molecular axis along the laser polarization vector, a feature of laser-molecule interactions that has been well studied [8]. We note that N_2 has a very low value of the empirical constant in the Hagena parameter [7], indicating that this molecule has low propensity for clustering; thus there is a component of unclustered N_2 molecules that also emerges from the skimmer.

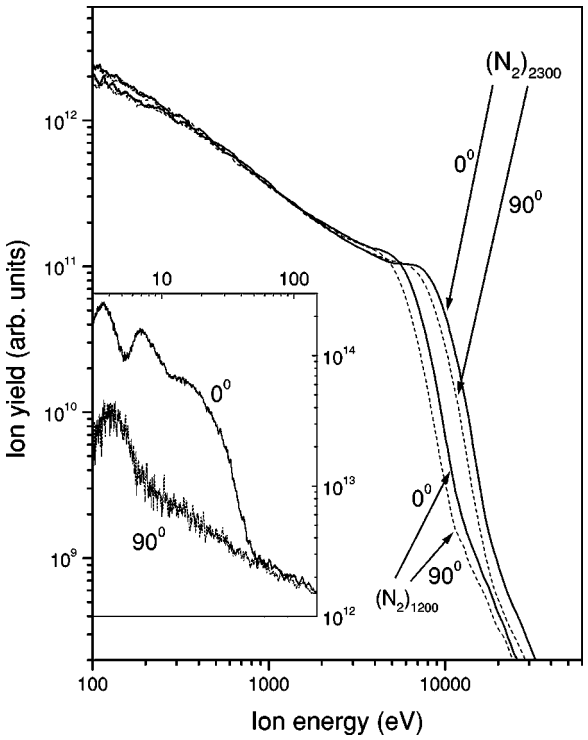

FIG. 1. Ion energy spectra for laser polarizations parallel (0°) and perpendicular (90°) to the TOF axis for $(N_2)_{2300}$ and $(N_2)_{1200}$ clusters at a laser intensity of 7×10^{15} W cm^{-2}. Note that for energies less than ~4 keV, the two spectra overlap; differences are only seen for high-energy components. Note that the "knee" energy shifts from 10 keV in the case of the larger cluster to 4 keV for the smaller one. The inset shows the asymmetry at very low energies ($\leqslant$40 eV). This is ascribed to spatial alignment of unclustered N_2 molecules.

Variation of cluster size does not result in alteration of the overall morphology of the energy distribution function but for a shift in the energy value at which the isotropic and anisotropic components meet. This point, which we refer to as the "knee," shifts progressively towards lower energies as the cluster size is reduced. The inset in Fig. 2 shows the experimentally measured ion spectra for $(N_2)_n$ at different

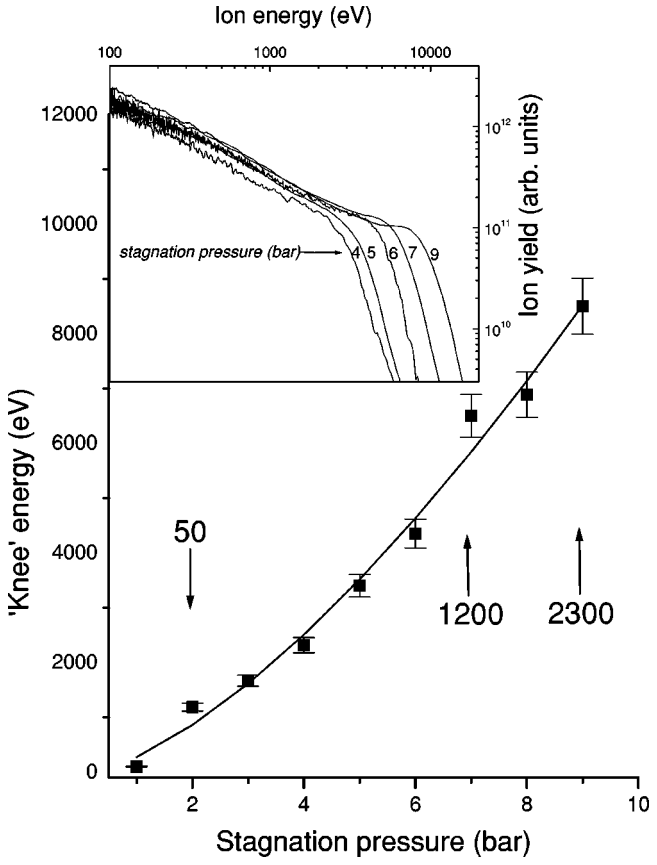

FIG. 2. Variation of "knee" energy with stagnation pressure P. The solid line is a fit to the measured data and indicates a $P^{\alpha}(\alpha = 1.5 \pm 0.2)$ dependence. Cluster sizes for a few values of P are indicated by the vertical arrows. The inset shows the ion energy spectra for laser polarization parallel to the TOF axis for different values of P, and demonstrates that the knee energy becomes smaller as the stagnation pressure is lowered.

pressures, when the laser light is polarized along the TOF axis.

The maximum value of ion energy, E_{max}, as $(N_2)_n$ explodes under Coulombic pressure is from ions that are on the outer surface of the spherical cluster comprising n_c atoms (molecules) and is given by

$$E_{max} \propto \frac{Q\bar{q}n_c}{R}, \tag{1}$$

where $\bar{q}$ is the average charge per atom in a cluster, of radius R, and Q is the charge on the ion that is detected. It is established that n_c varies as P^2 [6,7], where P is the stagnation pressure in the supersonic nozzle and, since n_c scales with cluster size as R^3, it is clear that the ion maximum energy, E_{max}, should scale as $P^{4/3}$. This analysis is based upon two assumptions: (i) at a given laser intensity, the average charge per atom does not appreciably change with cluster size and (ii) cluster expansion is such that the radius of the expanding cluster at the end of the laser pulse is proportional to the initial radius. Results of extensive numerical simulations of cluster explosions that Last and Jortner [9] have carried out provide ample justification for the validity of both assumptions. Figure 2 depicts the knee energy that we have measured for different values of P. For $(N_2)_n$ clusters, a fit to our data (solid line in Fig. 2) shows that the knee energy scales as P^{α}, where $\alpha = 1.5 \pm 0.2$, which is in consonance with expectations from the above analysis. Measurements on Ar_n clusters also show a similar behavior; the knee energy is found to scale as $P^{1.2\pm0.1}$. This brings to the fore the necessity of having to invoke an additional acceleration mechanism, over and above Coulombic pressure, to account for ions that possess energies in excess of the knee energy. It is also essential that the observed anisotropy is accounted for. By working with clusters of relatively small size we exclude the possibility of hydrodynamic pressure coming into play. In this connection it has not escaped our attention that ion energies at values less than the knee energy scale approximately quadratically with ion charge for both $(N_2)_n$ and Ar_n clusters, as would be expected in the Coulombic regime.

The anisotropy that is observed for the highest energy ions, beyond the knee energy, provides a clue as to the nature of this additional acceleration mechanism. As has been observed in earlier work on Ar_n clusters [3], and in numerical simulations of explosions of clusters such as Xe_{147} [10], the charge state of an ion can rapidly change in resonance with the applied optical field. The radial field due to the charged cluster and the applied laser field act in the same direction for half the optical cycle, and a high ionic charge state can be attained. But, in the next half cycle, these two fields oppose each other, and the net charge is reduced from its maximum value due to ion-electron recombination. This charge flipping gives rise to a net cycle-averaged force that serves to accelerate the ions. The simulations carried out by Ishikawa and Blenski for a small cluster, Xe_{147} [10], showed that such acceleration is much smaller for ions that are ejected perpendicular to the laser polarization vector. The acceleration due to this mechanism is ΔQE, where ΔQ is the charge change in each half cycle and E is the electric field.

To further evaluate the acceleration mechanism, we have carried out ion energy measurements also for circularly polarized light. Figure 3 shows measured spectra for $Ar_{40\,000}$ at 8×10^{15} W cm^{-2} intensity. We find that while the isotropic component remains unchanged for circularly polarized light, the asymmetric components have energies that are smaller than those obtained with the polarization vector parallel to the TOF axis, but larger than those when the polarization vector is perpendicular to the TOF axis. Numerical simulations [11] have shown that electron heating is independent of light polarization when the density is large and collisional bremsstrahlung dominates. Experimental evidence corroborates this in that the x-ray emission yields from clusters such as $Ar_{40\,000}$ are measured to be independent of the polarization state of the incident radiation [6]. Our observation that the isotropic component of the ion energy distributions is also polarization independent is consistent with this. Taken together, these facets of the laser-cluster interaction imply that the charge on the cluster, the electron density, and the electron temperature are all more or less independent of the polarization state. It is, therefore, reasonable to expect that value of Δq would be similar for both linear and circular

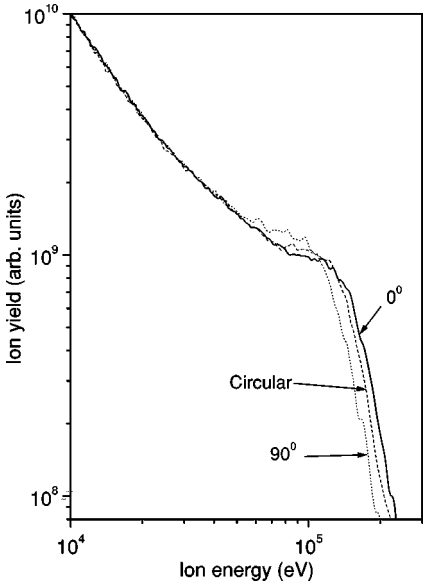

FIG. 3. Ion energy spectra obtained with $Ar_{40\,000}$ for laser polarizations parallel (0°) and perpendicular (90°) to the TOF axis, and also for circularly polarized light. The laser intensity was 8×10^{15} W cm^{-2}. The "knee" is identified as the energy at which a change of slope in the ion energy distribution function occurs, in the vicinity of 120 keV.

polarizations for clusters of the same size irradiated at similar intensities. Hence, the ratio of the mean energies of the asymmetric component should be proportional to the electric-field component along the TOF axis. We define the mean energy of the asymmetric component as

$$\langle E^{assym} \rangle = \frac{\int f(E^{assym}) E dE}{\int f(E^{assym}) dE}, \tag{2}$$

where $f(E^{assym})$ is the experimentally determined ion energy function beyond the knee. From data of the type shown in Fig. 3, it is possible to determine mean values of E^{assym} for experiments conducted with linear and circular polarizations. We have done this and analysis of the asymmetric component indicates that the ratio of mean energies of the asymmetric component obtained with linear and circular polarizations, R, is

$$R = \frac{\overline{E}^{assym}_{lin}}{\overline{E}^{assym}_{circ}} \tag{3}$$

which is equal to 1.38 ± 0.17. The error represents the standard deviation in the measured ratios for a number of measurements made at different values of laser intensity. This value of R correlates well with the value of 1.414 that is expected for the ratio of the electric-field component along the TOF axis for linear and circular polarization.

IV. CONCLUDING REMARKS

In summary, we have shown that disassembly, in the nonhydrodynamic regime, of atomic and molecular clusters yields ion energy distributions that have a low-energy isotropic component and a not-insubstantial asymmetric, high-energy component. From measurements of ion energy spectra for different cluster sizes we show that the isotropic component can be accounted for by purely Coloumbic considerations. The Coulomb limit has been experimentally established for clusters whose size varies from 50 monomers to 40 000. The high-energy component has ions whose energies are in excess of the Coulomb limit, and they are produced asymmetrically. The postulate of a charge-flipping mechanism was also invoked in earlier studies with Ar_n clusters [3] in order to qualitatively account for the observed anisotropy of high-energy ions. It was shown that for a unit change in ionic charge per optical cycle, additional energy of 30 keV is imparted to the fragment ions resulting from the cluster explosion. In the present study we have extended this earlier postulate by providing additional experimental evidence: from ion energy measurements made with linear and circularly polarized light, we show that the asymmetric component is essentially due to a charge-flipping acceleration mechanism of the type proposed by Ishikawa and Blenski following their Monte Carlo simulations of the disassembly of very small clusters. Although we have conducted experiments over a reasonably large range of cluster sizes, it isclearly necessary for three-dimensional Monte Carlo type of studies to be carried out in order to establish both the energies and the fraction of total ion yield that undergoes this type of acceleration. Exploitation of this additional acceleration mechanism might be of utility in future considerations of tabletop accelerators.

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