Strong-field ionization and Coulomb explosion of argon clusters by few-cycle laser pulses

D. Mathur,[1,*] F. A. Rajgara,[1] A. R. Holkundkar,[2] and N. K. Gupta[2]
[1]*Tata Institute of Fundamental Research, 1 Homi Bhabha Road, Mumbai 400 005, India*
[2]*Laser and Neutron Physics Section, Bhabha Atomic Research Centre, Mumbai 400 085, India*
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Energy distributions are measured for ions emitted upon Coulomb explosion of Ar_n clusters ($n = 400$–900) upon irradiation by intense three-cycle pulses (10 fs) of 800-nm laser light of peak intensity 5×10^{14} W cm^{-2}. With few-cycle pulses, there is insufficient time for the cluster to undergo expansion; this results in overall dynamics that are significantly different from those in the many-cycle regime. The peak ion energies are much lower than those obtained when 100-fs pulses of the same intensity are used; they are almost independent of the size of the cluster (over the range 400–900 atoms). Ion yields are measured to be larger in the direction that is perpendicular to the laser-polarization vector than along it. Model molecular dynamics calculations are used to qualitatively rationalize this unexpected anisotropy in terms of shielding by a spatially asymmetric electronic-charge cloud within the cluster.

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Intense few-cycle optical pulses are of considerable significance in the contemporary pursuit of attosecond pulses and in studies of light-matter interactions in the domain of ultrashort and strong fields [1]. Different strategies for few-cycle pulse generation have been demonstrated, such as hollow-fiber pulse (HFP) compression [2] and optical parametric amplification [3]. At present, HFP methods are the most widely adopted even though they present practical difficulties vis-á-vis alignment and low coupling efficiency. An alternate method based on filamentation in rare gases has also found recent utility [4], and pulses as short as 5 fs have been generated by using two-stage filamentation in argon gas. This is the strategy that has been adopted in our laboratory [5] to generate few-cycle pulses of intense 800-nm light, which have been used by us to probe the ionization and fragmentation dynamics of molecules [6] and to demonstrate a unimolecular bond rearrangement in H_2O on the time scale of a single vibrational period [7]. We now report application of such few-cycle pulses to studies of the strong-field dynamics of gas-phase clusters of Ar. Almost all hitherto-existing experimental work in the area of laser-cluster interactions in the strong-field regime has been carried out by using infrared laser pulses of 30-fs duration or longer (more than ten optical cycles). Our experiments have used intense pulses of 800-nm laser light in the few-cycle domain (10–35-fs pulse durations) wherein time-of-flight (TOF) spectrometry has been utilized to measure energy distributions of ions that are ejected upon Coulomb explosion of Ar_{400}–Ar_{900} clusters that become highly charged after irradiation by peak intensities of up to 5×10^{14} W cm^{-2}. The ionic products of Coulomb explosion of Ar clusters in the few-cycle domain are seen to have significantly lower energy than is the case upon irradiation with many-cycle pulses. Moreover, the dependence of ion yield on the polarization direction of the incident laser pulse is different in the few-cycle and many-cycle regimes: In the few-cycle experiments we have conducted, significantly more ions are produced when the incident polarization is in a direction that is perpendicular to the ion detection direction, in stark contrast to the results we obtain in the many-cycle (100-fs) domain [8]. We rationalize this unexpected result by means of model molecular dynamics calculations. Our experimental results throw new light on laser-cluster interactions in the ultrashort strong-field domain and open opportunities for further work that is qualitatively new.

The course of the last 15 years has seen the physics that governs the interaction of laser fields with large gas-phase clusters develop into a rich and vibrant subset of research on the behavior of matter in strong optical fields, and several reviews have been published [9–12] that provide a cogent overview of different insights that have been gained. A major physics question that remains open after 15 years worth of sustained experimental and theoretical work relates to the mechanism by which energy transfers from the optical field to the cluster. It is primarily electrons in the cluster that are the direct absorbers of optical energy; the absorbed energy is collisionally redistributed within the cluster so as to produce energetic ions, electrons, and, at higher densities, intense incoherent radiation. The level of ionization and the mean energy of the electrons and ions are significantly higher than those obtained in strong-field ionization of single atoms and molecules. The hot nanoplasma that is created within the irradiated cluster undergoes spatial expansion and breaks up in a few picoseconds, which yields ions with kinetic energies that can be as high as a few megaelectron volts [13], up to 5 orders of magnitude larger than the energies obtained upon Coulomb explosion of single multiply charged molecules [14]. Our measurements on cluster disassembly, which leads to ion production, were made in the hitherto-unexplored temporal regime (10 fs) that is substantially much shorter than typical cluster expansion times.

According to prevailing wisdom, collisional absorption (inverse bremsstrahlung) is the most likely energy-transfer mechanism, which involves the transfer of energy from electrons to other particles in a series of inelastic collisions within the cluster nanoplasma that is created upon initial field-induced inner ionization. In cluster parlance, inner ionization refers to ejection of electrons from the individual Ar atoms that constitute the cluster in our experiments. Such electrons are quasifree: They are not bound to any specific Ar atom, but they are spatially constrained within the cluster by the

*atmol1@tifr.res.in

Coulombic charge of the positive ions. It is these spatially constrained electrons that are subjected to heating by the oscillating optical field and the ensuing collisions with plasma constituents that can be thought to be responsible for the bulk temperature of the plasma. As further inner ionization occurs, the plasma expands due to hydrodynamic and Coulombic forces, and its density becomes less. The buildup of ionic charge within the expanding cluster eventually leads to its explosion, whereupon energetic charged particles are emitted. There are alternative (noncollisional) mechanisms, such as resonance absorption [15] and vacuum (or Brunel) heating [16] that might contribute to energy transfer. In this paper, we discount effects of these processes by taking recourse to few-cycle pulses with peak intensities that lie below 10^{15} W cm^{-2}. This intensity regime allows us to ignore resonance absorption; our temporal regime allows us to satisfactorily deal with a less-addressed facet of the collisional mechanism, namely, the role played by enhanced ionization (EI) as a possible contributor to energy absorption by the cluster. The role of EI in clusters has not been explicitly addressed in any experimental study thus far [9]. We effectively switch off EI by the simple expedient of using laser pulses that are too short for significant nuclear motion to occur.

Subsequent to the submission of this paper, Skopalová *et al.* [17] have also reported on experiments carried out on Ar and Xe clusters with 10-fs-long pulses that demonstrate anisotropic ion emission, with more ions emitted perpendicular to the polarization direction than parallel to it. Moreover, these studies illustrate how such anisotropy appears to depend on pulse duration, with the anisotropy reverting to the usual situation (preferential ion emission along the polarization direction [8]) for longer pulse durations.

In our experiments, laser pulses (800-nm wavelength, 1-kHz repetition rate, 1-mJ peak energy) as short as 10 fs were generated by using two-stage filamentation in Ar [5]. Generation of gas-phase clusters was by standard techniques that have been used in most recent experiments, which include those from our laboratory [8,18]: The mean size of our Ar clusters was varied from Ar_{400} to Ar_{900} by controlling the stagnation pressure behind a high-pressure conical gas nozzle (0.5-mm diameter). The laser beam, the cluster beam, and the TOF axis were mutually orthogonal to each other in an oil-free ultrahigh-vacuum environment (base pressure $\leqslant 10^{-9}$ Torr, stagnation pressure 2–10 atm). Standard TOF methods were used to mass spectrometrically separate the ionic-charge states obtained upon cluster explosion. Details of the apparatus has been published previously [8,18]. A simple transformation of the measured ion TOF spectra enabled temporal information to be mapped into energy spectra [18], typical examples of which are shown in the following.

Figure 1 shows results obtained when Ar_{400}, Ar_{750}, and Ar_{900} clusters were irradiated with pulses of 10-fs duration, with 5×10^{14}-W cm^{-2} peak intensity. We note three different facets of these energy distributions that distinguish them from those that are obtained in measurements made with many-cycle pulses of the same intensity [9].

First, we note that maximum ion energies for all these clusters lie in the range 2.7–3.0 keV: There is little variation with cluster size. This is in marked contrast to size-dependent ion energies that have been measured with longer pulses.

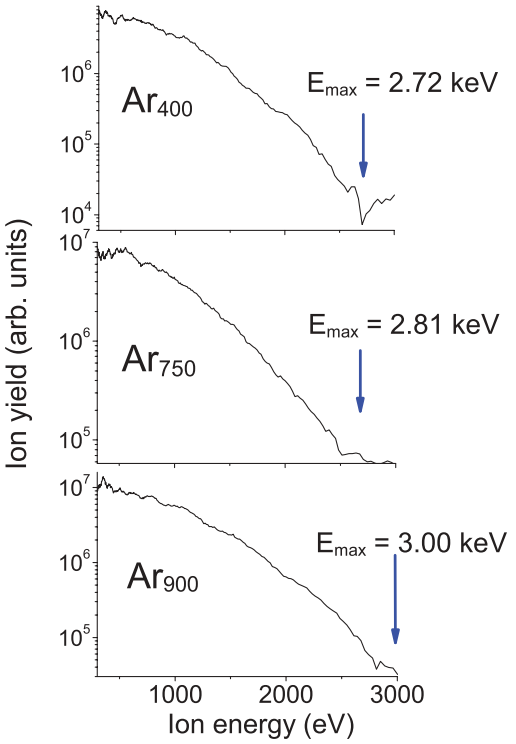

FIG. 1. (Color online) Energy distributions of ions obtained upon irradiation of Ar clusters with three-cycle (10-fs) pulses of 800-nm laser light of intensity 5×10^{14} W cm^{-2}. The laser-polarization vector was perpendicular to the ion-detection axis. Ion yields were several fold smaller when the laser-polarization vector was parallel to the ion-detection axis.

Second, the shapes of the energy-distribution functions are noteworthy in that they differ significantly from shapes that are obtained in the many-cycle (100-fs) temporal domain [12]. The third facet is possibly the most unexpected: Our data for Ar clusters show a strong anisotropy of the ion-energy spectrum with respect to the laser-polarization vector such that the ion signal that we detected perpendicular to the laser-polarization vector was severalfold higher than in the parallel direction. This is also in marked contrast to the findings in many-cycle experiments (see Ref. [9] and references therein).

Earlier work [8,18] has established that ion-energy distributions obtained with 100-fs pulses comprise two distinct components. One component results in maximum values of ion energies that are consistent with what would be expected on the basis of the Coulomb law applied to a charged spherical cluster of a given size. This component is isotropic with respect to the incident-polarization vector. Additionally, there is a second higher energy ion component whose maximum energies extend well beyond the Coulomb limit. This high-energy component exhibits anisotropy with respect to the laser-polarization vector [8,18]. The two components are separated in measured energy-distribution functions by a kneelike feature that is missing in our few-cycle data (Fig. 1). The angular anisotropy experimentally observed with 100-fs pulses was subsequently theoretically rationalized [9] to be a consequence of polarization-induced effects that arise from the phase difference between the optical field and the oscillations of the negatively charged cloud generated within the cluster by

inner-ionized electrons. Such effects give rise to a flipping of charges at the two poles of an initially spherical cluster [8,18], with the poles aligned along the laser's E vector. Our data show that such charge flipping is of no consequence when cluster irradiation is by a three-cycle pulse, as there is insufficient time for significant phase difference to be accumulated. We discuss this more in the following; but, prior to that, we note the facet of our results which relates to the size independence of maximum ion energy in our three-cycle experiments. This is of interest from the viewpoint of implications which arise from calculations reported by Breizman and co-workers on the electron response and ion acceleration in clusters by using a nonlinear approach [19]. These calculations were carried out by using two methods to describe the incident-laser field and addressed cluster size issues in the following manner. For optical fields that are taken to be oscillating, we compute, by using the treatment of Breizman *et al.*, that ~2% of ionized electrons actually leave Ar_{900}, while only ~10% leave Ar_{400}. Consequently, there is relatively little change in the density of inner-ionized electrons as cluster size increases from 400 atoms to 900 atoms. Consequently, the extent to which these clusters are collisionally heated is also not strongly affected by size; and, hence, we surmise that there is little dependence of maximum ion energies on cluster size. This, indeed, is consistent with our observations. On the other hand, the treatment of Breizman *et al.* [19] also allows the optical field to be treated as a static field. In that case, we compute that a much larger fraction of electrons leaves the cluster, which varies between 53% for Ar_{400} to about 35% for Ar_{900}; this larger variation would manifest itself in the extent of collisional heating which is size dependent and, consequently, the maximum ion energies are dependent on cluster size. Our results are, indeed, consistent only with those that are theoretically expected when an oscillating optical field is made to act on cluster electrons in the theoretical treatment of Breizman *et al.* [19]. An oscillating field is seen to be an apt description in the few-cycle domain, whereas a static field might suffice in the longer-pulse many-cycle domain.

We now address the third significant facet of our results, namely, the very strong anisotropy of the ion-energy spectrum with respect to the laser-polarization vector. The ion yield that was measured was strongly anisotropic, with a very significant increase in yields when the ion signal was detected perpendicular to the laser-polarization vector. A typical example of a TOF spectrum of Ar^+ ions from Ar_{700} is shown in Fig. 2, where measurements for orthogonal polarizations were made for the same number of laser shots, with an ion-extraction field of 200 V cm^{-2} applied across the laser-cluster interaction zone and with all experimental conditions (laser intensity, stagnation pressure) kept constant. Similar effects were obtained for $Ar^{2+,3+,4+}$ ions. It is clear that ion emission in the direction that is perpendicular to the laser polarization is much stronger than for parallel polarization, in stark contrast to earlier measurements of the angular distributions of energetic ions emitted upon explosion of Ar and Xe clusters irradiated with 100-fs pulses [8,18]. We attempt to rationalize these observations in terms of differing shielding processes that occur in the few-cycle and many-cycle temporal regimes. We have taken recourse to model calculations of electron dynamics on ultrashort time scales by using a molecular dynamics (MD)

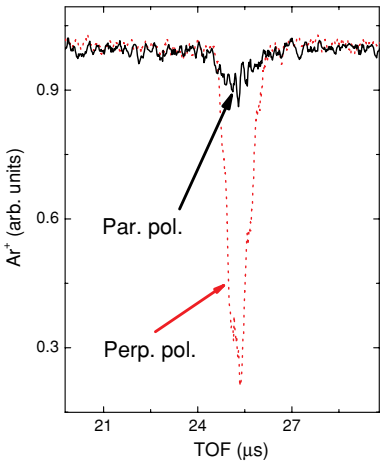

FIG. 2. (Color online) Raw TOF spectra of Ar^+ ions from Ar_{700} upon irradiation by 10-fs laser pulses whose polarization vector was aligned parallel (Par. pol.) and perpendicular (Perp. pol.) to the spectrometer axis.

approach of the type described by Petrov and co-workers [20]. For illustrative purposes, here, we consider a single Ar cluster of radius R_0 whose center is taken to lie at the origin of the coordinate system ($x = y = z = 0$). The number of atoms N, which comprises the cluster size is $N = (R_0/R_W)^3$, where R_W is the Wigner-Seitz radius. For $R_W = 2.4$ Å, a single 30-Å cluster comprises 1950 Ar atoms. The presence of neighboring clusters is accounted for by imposing periodic boundary conditions on the faces of our simulation cubical box with each side equal to the intercluster distance R_{IC} (in the present case, $R_{IC} = 20R_0$). Cluster ionization is implemented in Monte Carlo fashion, by using the oft-used Ammosov-Delone-Krainov tunnel ionization formula [21] applied to each ion and atom after each time step Δt for which we compute the force due to the optical field and the Coulomb force due to the presence of other charged particles. Each particle is then advanced to its new position according to its

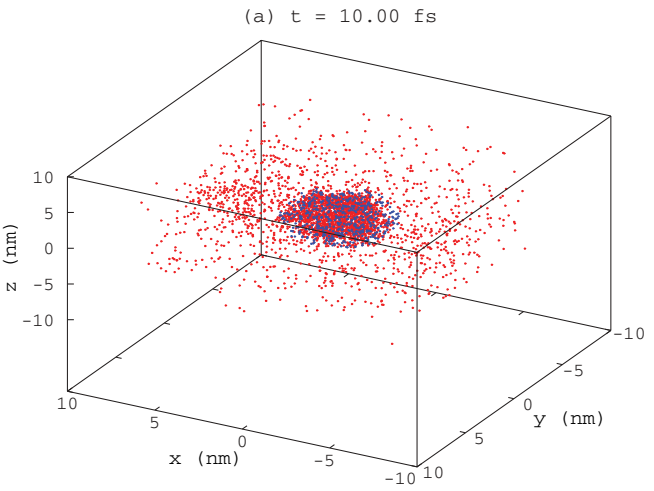

FIG. 3. (Color online) A snapshot of the asymmetric electron (dense blue dots) and ion (spread-out red dots) spatial profiles in a 30-Å Ar cluster after irradiation by a 10-fs laser pulse. The laser-polarization vector is aligned along the x axis.

relativistic equations of motion. In Fig. 3, we focus on the spatial distribution of electrons produced within the cluster upon irradiation by a 10-fs pulse. The Coulomb explosion occurs between 20 and 40 fs; and, prior to it, the buildup of the electronic-charge cloud is seen to be asymmetric, with alignment of the asymmetric electronic-charge cloud along the direction of the laser-polarization vector.

This asymmetric electronic-charge cloud shields ions within the core of the cluster such that the extent of shielding is spatially inhomogeneous: The polar regions of the spherical cluster (in the direction of the laser-polarization vector) are much more effectively shielded than the equatorial regions. Consequently, when the ionic-charge cloud in the core builds up sufficiently to result in a Coulomb explosion, the products of this explosion manifest themselves more prominently along the equatorial region than in the polar regions. This is precisely what we observed, with a significant diminishing of ion yields in the laser-polarization direction compared to what is obtained in the orthogonal direction (Fig. 2). The indications that emerge from our MD calculations can only be regarded as rough guideposts to rationalization of the observed ionic anisotropy. It is clear that more elaborate calculations need to be undertaken in order to properly account for the shielding effects of the electronic-charge cloud within the cluster. In long-pulse experiments, such shielding is of little consequence as the charge-flipping process referred to earlier dominates the ion dynamics and leads to the opposite type of asymmetric emission of Coulomb explosion products, more along the polarization direction than perpendicular to it.

Now, we consider, in more detail, the dependence of the ion-energy distribution functions on pulse duration. Figure 4 shows energy spectra measured for pulses of 10-fs and 35-fs duration. The pulse energy was appropriately compensated in these two measurements to ensure that the peak intensity experienced by Ar_{900} clusters was the same. The maximum energy obtained with 35-fs pulses is 3.9 keV, compared to 3.15 keV in the case of 10-fs pulses. The theoretical treatment of Breizman *et al.* [19] predicts that, for very short pulses, the ion energy will scale as τ^2, where τ denotes the laser-pulse duration. It is clear that this scaling is not observed in the present experiments. It is also noteworthy that the symmetric shielding implicit in the model of Breizman *et al.* [19], as utilized in the recent work of Skopalová *et al.* [17], may not be a robust enough description of the short-time evolution of the electronic-charge cloud within Ar clusters: Our MD simulations exhibit a pronounced anisotropic shape of the charge cloud on time scales of 10 fs.

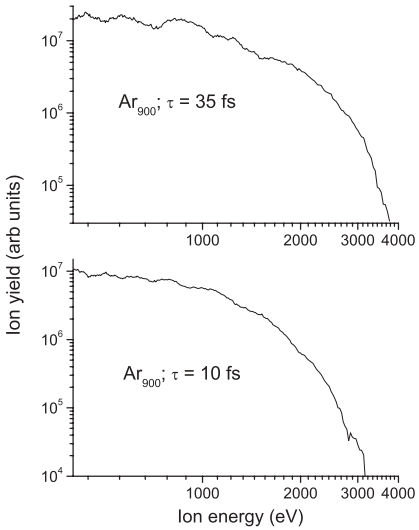

FIG. 4. Ion-energy spectra measured by using pulses of 10-fs (top panel) and 35-fs (lower panel) duration, both of peak intensity 5×10^{14} W cm^{-2}.

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