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Dynamics of atomic clusters in intense optical fields of ultrashort duration#

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Abstract. Intense laser pulses have been generated that last for only 10 fs, long enough to accommodate only 3 optical cycles of 800 nm light. Upon focussing such pulses, intensities in the 10^{15} W cm^{-2} range are readily generated. At such intensities, the magnitude of the optical field begins to match intra-atomic Coulombic fields. Consequently, exposure of atoms and molecules to such intense pulses inevitably leads to single and multiple ionization. We report here results of experiments that we have conducted that involve irradiation of gas-phase $Ar_{15,000}$ clusters by such intense, few-cycle laser pulses. The clusters become multiply ionized and undergo Coulomb explosion, giving rise to ejection of fast Ar-ions. Results show that the strong-field dynamics in the few-cycle domain differ significantly from those that occur in the longer pulse (>30 fs) regime. Manifestations of these differences are presented in the form of angle-dependent ion energy and ion yield functions.

Keywords. Atomic clusters; Coulomb explosion; few-cycle laser pulses; strong fields; cluster dynamics.

1. Introduction

A number of scientific and technological developments are responsible for the resurgence of interest in studies of light-matter interactions, particularly of how very intense light interacts with matter. The interest stems from the fact that very intense light has associated with it very strong fields, and the dynamics that ensue when matter is exposed to such fields are of both fundamental and applied interest. It is useful to begin by defining the terms 'strong' field and 'intense' light with reference to some physically established standard. The Coulombic field that is experienced by an electron in the 1 s orbital of the hydrogen atom is one useful benchmark. It has a value of $\sim 10^9$ V cm^{-1}. The intensity of light that would give rise to such a field is 10^{16} W cm^{-2}. Light of such intensity is readily generated using pulsed lasers, like a Ti:sapphire laser that emits 800 nm light, typically with pulse durations of 100 fs or thereabouts. It is not difficult to achieve remarkably high photon densities, of the order of 10^{36} photons s^{-1} cm^{-2}, by focusing light from commercially available femtosecond lasers. Such photon densities mediate the essentially nonlinear nature of light-matter interactions, where matter comprises atoms, molecules and, for the purposes of the work that we describe here, large gas-phase clusters of atoms.

The mesoscopic nature of cluster dynamics in intense laser fields exhibits features that are unique to clusters, not observed either in the interaction of individual atoms and molecules exposed to fields of similar magnitude and duration, nor in the case when the irradiated matter is in the condensed phase. For example, the extraordinarily high degree of efficiency with which clusters absorb the incident laser energy finds no analogue in other forms of matter. Energy deposition rates of up to 240 mW *per atom* have been measured[1] in clusters comprising several hundreds of thousands of atoms. The electrons within the cluster are the direct absorbers of such energy, which is rapidly redistributed as intense incoherent radiation and emission of energetic particles. The level of ionization and the mean energy of the electrons are both significantly higher than that which occur upon ionization of isolated (gas-phase) atoms and molecules. Multiple ionization implies the existence of a large number of electrons and ions; in the context of an atomic cluster, it also implies the existence of a hot plasma of nanometre dimensions. The hot, highly-ionized cluster expands due to Coulombic and hydrodynamic pressure, which eventually breaks up in a few picoseconds. This rapid disassembly results in the emission of ions with kinetic energies that can be as high as a few MeV,[2] energy of five orders magnitude is obtained when multiply charged molecules undergo Coulomb explosion.[3]

#Dedicated to Prof. N Sathyamurthy on his 60th birthday
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The distinguishing facet of an exploding cluster being the source of very energetic charged particles has provided one important motivation for research into cluster dynamics in strong fields in recent years. From the viewpoint of basic science, we note that clusters are intermediate to atoms and small molecules on the one hand, and to solids on the other. The transition from single atom behaviour to bulk properties is complex enough to merit investigation for its own sake. Upon irradiation of clusters that are not too large, a substantial fraction of electrons ionize and leave the cluster early in its break-up, leaving behind a hot, non-equilibrium and ionized plasma, the evolution of which is strongly influenced by the optical field, sometimes leading to the formation of a radiative blast wave[4] of the type that is of interest in astrophysics; cluster plasmas offer one of the few avenues available for studying such energetic wave phenomena in the laboratory. From an applied viewpoint, clusters offer tantalizing possibilities of developing table-top accelerators and as sources of extreme ultraviolet (EUV) radiation for lithography, the next generation of chip manufacturing technology. Apart from incoherent radiation, clusters also show promise as a source for high-harmonic generation, wherein extremely non-linear processes give rise to coherent radiation at large multiples of the laser frequency.[5] The interaction of laser pulses from table-top lasers with deuterium and deuterium-rich clusters has also made it possible to study hot-plasma fusion in small laboratories, which was hitherto restricted to very large national and international facilities.[6] The fusion process also holds promise as a source of mono-energetic and short-pulse neutrons for medical and material science studies.[7]

Various theoretical models have been developed to rationalize the evolution of the cluster under intense field irradiation; however, none can adequately explain the experimentally observed features that presently drive research in this area. Most atoms in a cluster undergo tunnel ionization at the leading edge of the incident laser pulse. As the ionized electrons are removed from the cluster, a positively charged core is left behind that gives rise to an increasing potential barrier to further removal of electrons. Hence, individual atoms in the cluster are ionized ('inner ionization') but the ionized electrons may still be trapped within the cluster volume, thereby precluding 'outer ionization' of the cluster as a whole. Whether the potential barrier is sufficient to retain a large fraction of electrons within the cluster volume or not is a major bone of contention between the major theoretical models. In the hydrodynamic expansion model, retention of most of the electrons is assumed, giving rise to a spherically symmetric nanoplasma. Such retained electrons absorb energy from the optical field by collisional inverse bremsstrahlung; the hot electron plasma expands due to hydrodynamic pressure, and transfers energy to the ions. The expansion velocity of the plasma is determined by the plasma sound speed, and since the ion and electron charge clouds expand at the same speed, the ions becoming significantly 'hotter' than the electrons. On the other hand, the Coulomb explosion-ionization ignition model assumes that ionized electrons are ejected from the cluster volume and, consequently, there is a build-up of positive charge within the cluster that gives rise to a radial field which may become large enough to drive further ionization at the surface of the sphere. The removal of these electrons from the surface further enhances the radial field, and 'ignites' further ionization. The cluster then explodes due to the Coulombic repulsion between the positively charged ions.

2. Clusters in strong optical fields

It is clear that in the course of the last decade or so, various experimental studies of how clusters interact with intense laser fields have brought to the fore several

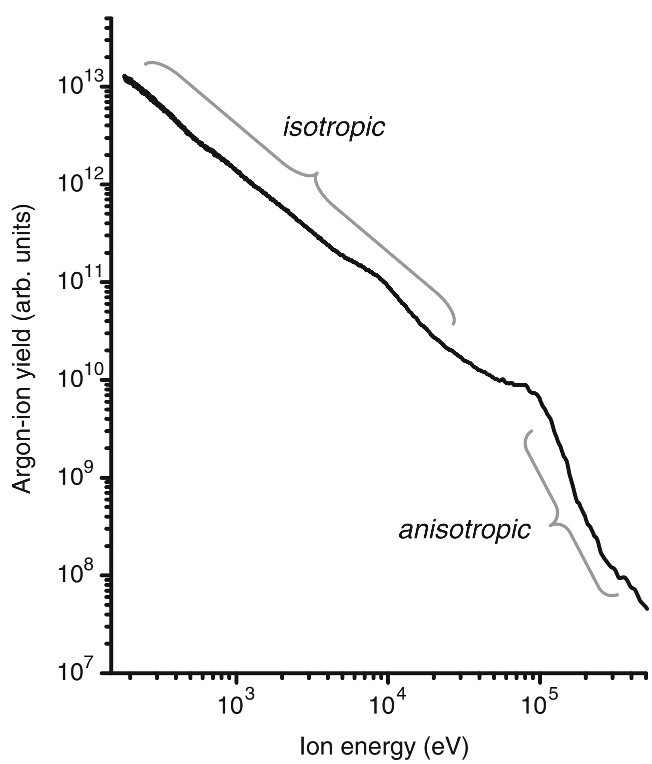

Figure 1. Typical energy spectrum of ions emitted upon explosion of $Ar_{40,000}$.

unique properties that clusters possess compared with atoms, molecules and bulk matter, properties that have been cogently reviewed in recent publications.[8–11]

Figure 1 shows a typical energy spectrum, measured in our laboratory, of Ar-ions emitted upon explosion of a large cluster, $Ar_{40,000}$, upon irradiation by laser pulses of intensity 8×10^{15} W cm^{-2}. The pulse duration in these experiments was 100 fs. The spectrum is typical of ones that have been reported earlier[12] and can be considered archetypal. Note that (i) maximum energies approaching 1 MeV are obtained for $Ar_{40,000}$, and (ii) the shape of the energy distribution has a knee-like structure, in the region of 100 keV in the case depicted in figure 1, that delineates two regions of different slopes. This shape has been found to be typical of most energy distribution functions obtained from a variety of clusters and affords an experimental handle that enables the testing of the efficacy of different theoretical models in properly describing laser-cluster dynamics in the strong field regime. For both atomic[12] as well as molecular clusters,[13] experiments have established that the highest energy ions are anisotropically emitted: for incident light that is linearly polarized, maximum ion intensity is obtained in a direction parallel to the laser polarization vector. The polarization dependence is a consequence of changes in the charge states at the poles of a spherical cluster in half a cycle of the laser field (the poles are defined with respect to the direction of the laser polarization vector). Consider two consecutive optical half cycles (see figure 2); in the first half cycle, the 'bottom' or 'South' pole may be depleted of electrons due to the combined action of the laser field and the radial electric field that arises from the charged nature of the cluster, with ions at the surface more likely to lose electrons than those in the core. In the second half cycle, the electrons are driven back towards the bottom pole, leaving the 'North' pole at the 'top' highly charged. This alternation, or charge flipping, results in a net cycle averaged force on the ions at the poles. Electron-ion collisions within the spherical plasma also contribute to the charge flipping such that, after one optical cycle, the net charge at the 'top' of the sphere is not the same as that at the 'bottom'. 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.[12,13]

The manifestation of such charge-flipping and consequently, the polarization dependent highest energy component of the ion energy distribution, in energy spectra is also shown in figure 2. The energy regime below the knee energy yields ions isotropically while those ions that possess energies in excess of the knee energy (the maximum Coulombic energy) exhibit an anisotropy. From the perspective of an 'ion source' fortunately, it is the most energetic ions that are most amenable to formation of an ion beam as far as considerations of beam parameters like brightness and angular divergence are concerned.

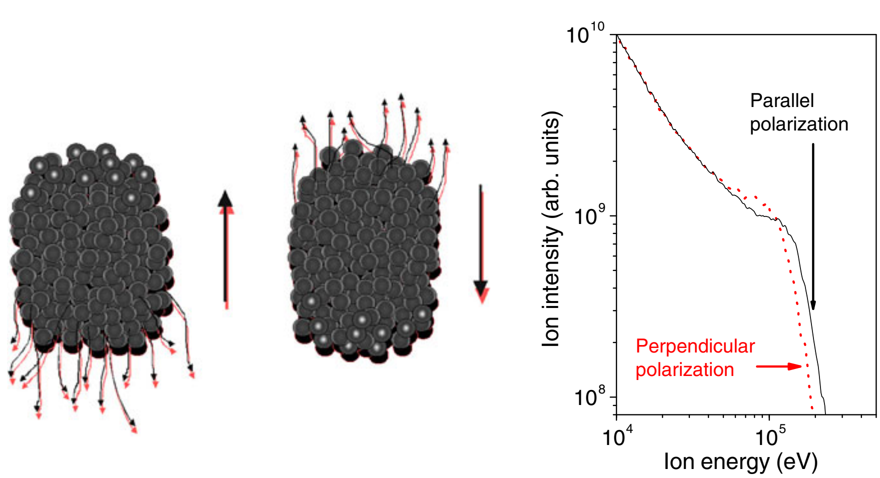

Figure 2. Left panels: Schematic representation of charge flipping and its experimental manifestation in the ion energy spectrum (right panel) obtained when $Ar_{40,000}$ explodes upon irradiation at an intensity of 5×10^{15} W cm^{-2}. The experiment was conducted using 100 fs long pulses of 800 nm light. The thicker vertical arrows denote the direction of the optical field, which changes every half cycle. The thinner arrows at the top and bottom of the cluster denote ionized electrons.

3. Ultrashort pulses

Thus far the discussion of cluster dynamics in strong fields has taken no explicit cognizance of the pulse duration responsible for the strong field. Exploration of the temporal facet of the dynamics has recently become feasible due to development in methods of generating intense laser pulses of only a few femtoseconds duration, long enough for only a few optical cycles. The first set of results have already yielded some surprises.[14–16]

Generation of intense, few-cycle optical pulses has become important in the contemporary pursuit of attosecond pulses and in studies of ultrafast dynamics and light-matter interactions in general. In most laboratories it is the hollow-fiber pulse compression technique[17] that is used to generate few-cycle pulses although an alternate method based on filamentation in rare gases has recently begun to also find utility.[18] It is the latter technique that has been adopted in our laboratory[19] to probe the ionization and fragmentation dynamics of molecules with four-cycle pulses,[20,21] and to demonstrate a unimolecular bond rearrangement in water on the timescale of a single vibrational period.[22] Specifically, we focused 40 fs laser pulses

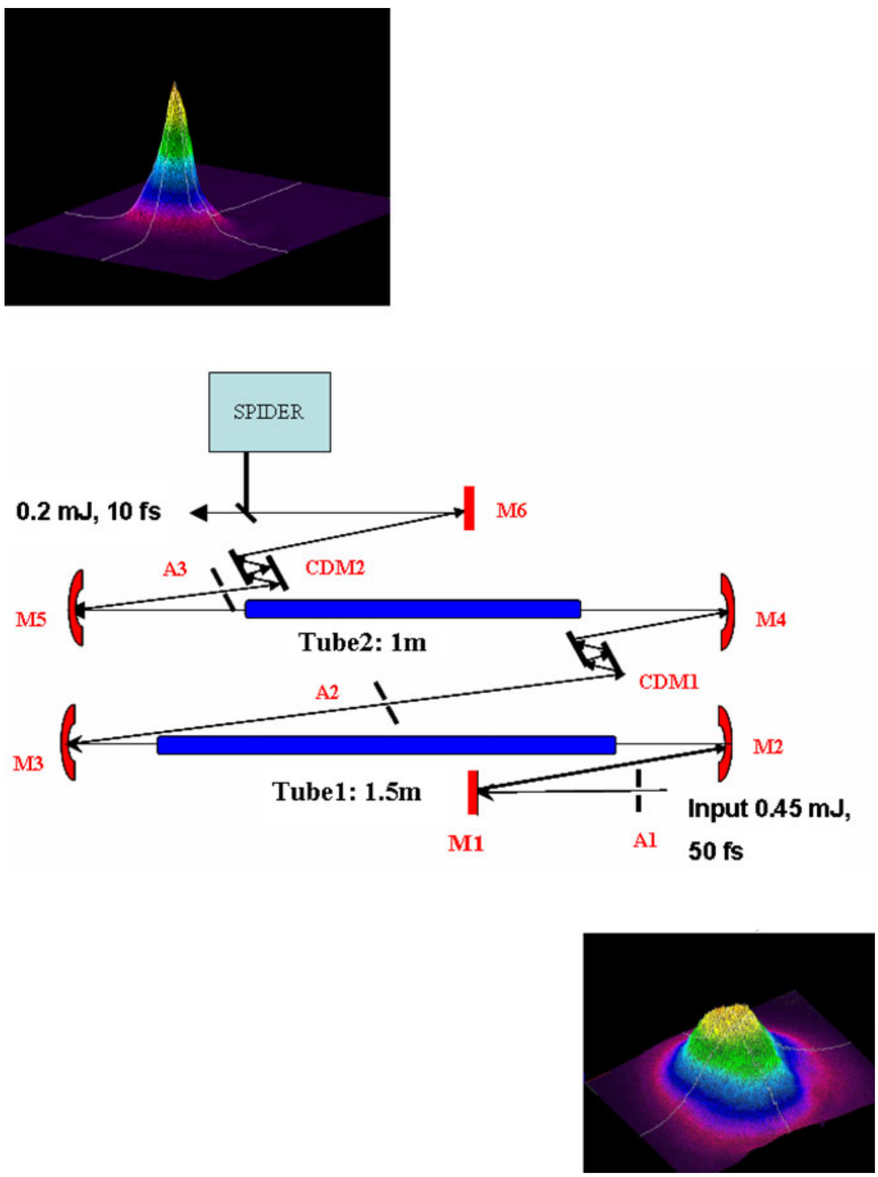

Figure 3. Schematic representation of our filamentation-based method of generating few-cycle pulses. The beam profiles at the lower and upper portions of the figure represent the input and output profiles, respectively (see text).

(800 nm wavelength, 1 kHz repetition rate, 1 mJ peak energy) into two 1 m long tubes filled with Ar-gas (typically at 1 atm pressure) using two-stage filamentation (figure 3).[19] Following filamentation in Ar, the white light that is generated was temporarily re-focused with two sets of chirped dielectric mirrors so as to generate pulses as short as 7–10 fs (2–3 optical cycles). Note how the laser beam profile at the output of our compression system is a marked improvement over the input beam profile. Characterization of the few-cycle pulses we thus generate is by means of a spectral interferometric technique (details of our generation methodology and diagnostics have been recently published).[19]

Generation of gas-phase clusters in the present series of experiments was by supersonic expansion techniques that have been used by us in earlier studies[12] with longer (100 fs) pulses: the mean size of our Ar-clusters in the series of experiments that we report upon here was varied from Ar_{400} to $Ar_{15,000}$ by controlling the stagnation pressure behind a high-pressure conical gas nozzle (0.5 mm diameter). The laser beam, cluster beam, and TOF axis were mutually orthogonal to each other in an oil-free, ultrahigh vacuum environment (base pressure $\sim 10^{-9}$ Torr, stagnation pressure 2–10 atmospheres). Standard time-of-flight (TOF) methods were used to mass spectrometrically separate the ionic charge states obtained upon cluster explosion. A simple transformation of the measured ion time-of-flight spectra enabled us to map temporal information into energy spectra (figure 1).[13]

4. Results and discussion

Very recent applications of intense, few-cycle pulses to clusters of argon and xenon have yielded some interesting and, initially surprising results. Hitherto-existing experimental work in this area was overwhelmingly carried out using laser pulses whose pulse duration (>30 fs) is long enough to accommodate 10 optical cycles or more. Experiments in the *few-cycle* domain have revealed that energy distributions of ions that are ejected upon Coulomb explosion of highly charged $Xe_{500-25,000}$ and $Ar_{400-900}$ clusters yield maximum energies that are very significantly lower than those obtained when longer pulses of the same intensity are used.[14,15] This is a consequence of the few-cycle temporal regime not allowing the irradiated clusters sufficient time to undergo significant expansion: the nuclear degrees of freedom are effectively ‘frozen’ when pulses of the order of 10 fs are used. The other significant difference that has been reported is that ion yields in the few-cycle experiments are polarization dependent in different fashion to what is indicated in the typical long-pulse data shown in figure 2. In the few-cycle domain, ion yields have been discovered to be significantly larger when the polarization is perpendicular to the detection axis than along it.[15,16] This unexpected behaviour is qualitatively rationalized in terms of a spatially anisotropic shielding effect induced by the electronic charge cloud within the cluster.[15] The results that we present in the following are an extension of these recent studies: we report energy and angular data obtained upon irradiation of larger Ar clusters. The present work helps shed new light on laser-cluster dynamics in the ultrashort, strong-field domain and opens opportunities for further work that is qualitatively new.

Figure 4 shows a typical ion energy spectrum measured when $Ar_{15,000}$ are exposed to 10 fs long pulses of peak intensity 8×10^{14} W cm^{-2}. Two noteworthy features of the spectrum are: (i) the absence of a knee-like structure of the type that is routinely observed in longer-pulse ($\sim$100 fs) experiments and (ii) the polarization-independent shape of the energy distribution function. The first feature appears consistent with the recently reported observation that the maximum ion energy measured with 10 fs pulses is distinctly lower than that obtained when the same cluster is exposed to 100 fs pulses of the same peak intensity. As noted earlier, this is a consequence of there being insufficient time for adequate heating to occur of inner-ionized electrons within the cluster: the resulting nanoplasma is, as a result, less ‘hot’ in the ultrashort regime than it

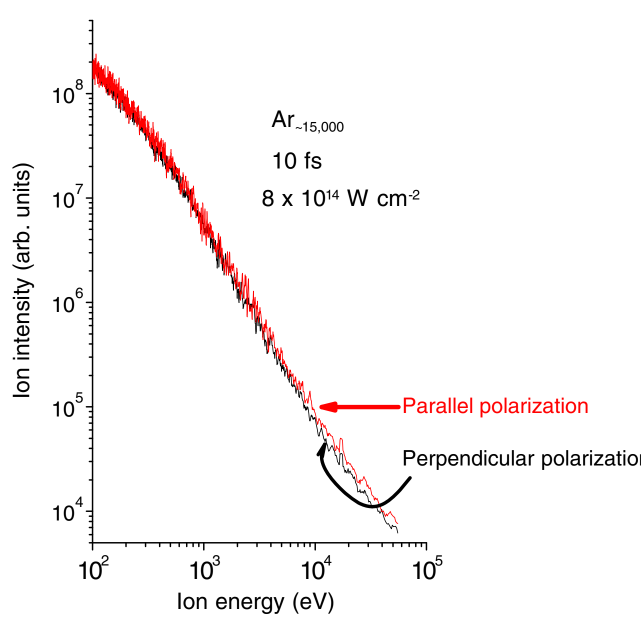

Figure 4. Distribution of kinetic energies of ions ejected upon explosion of $Ar_{15,000}$ clusters.

would be in the 100 fs regime. Assuming the cluster to be spherical in shape, a simple calculation reveals that the maximum energy in the spectrum in figure 4 is not inconsistent with the Coulomb explosion energy that would be expected for a cluster of 15000 *Ar*-atoms with an overall charge state of 8+. There is simply not enough time for any charge flipping to occur to give the ions an additional energy 'kick' over and above the Coulomb explosion energy: hence the absence of a knee that is a signature of extra-Coulombic energy.

The second feature of the energy spectrum shown in figure 4, namely the polarization independence of the ion energies, is a new result that has not been reported earlier but one that is consistent with our discussion thus far. As the energies of the exploding ions are fully consistent with a pure Coulomb explosion mechanism, with no extra charge flipping component, then the angular distributions of the ions would also be expected to follow Coulombic principles. For a spherically symmetric cluster, the explosion would eject ions in isotropic fashion, and this is exactly what the energy data in figure 4 represents. It is, however, of interest to note that as far as ion yield are concerned, there is still an asymmetry. Figure 5 shows how the yield of Ar^+ ions exhibits strong polarization dependence but one that is directionally opposite to what is observed in long-pulse experiments. Our ion yield data (figure 5) is in accordance

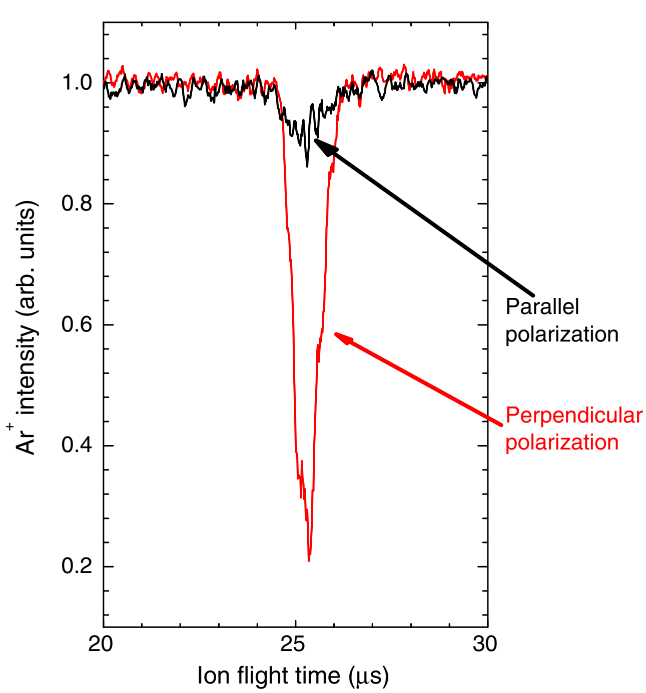

Figure 5. Raw time-of-flight data showing the yield of Ar^+ ions obtained with two orthogonal laser polarization directions.

with recently-presented data for Ar- and Xe-clusters of smaller size.[14–16]. Molecular dynamics calculations carried out by Gupta and coworkers for Ar-clusters[15] have succeeded in rationalizing this unexpected angular dependence in terms of asymmetric charge shielding effects within the irradiated cluster. Such asymmetric charge shielding plays an important role in the few-cycle domain; its effect is neutralized once the charge flipping process depicted in figure 2 comes into play. It would clearly be of interest to carry out more comprehensive molecular dynamics studies to determine what properly accounts for the differing angular behaviour of ion yield and ion energy functions in the few-cycle domain.

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