Physica Scripta. Vol. T80, 35–39, 1999

The Generation of High Energy Ions by Photo-Induced Dissociation of Atomic Clusters

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Received October 12, 1998; accepted October 16, 1998

PACS Ref: 36.40, 52.50.J, 07.85.F, 32.30.R

Abstract

The interaction of an intense laser pulse of sub-picosecond duration with an atomic cluster larger than a few hundred atoms can be extremely energetic. The high local density within the cluster together with a dynamic dielectric resonance in the expanding cluster microplasma greatly enhance coupling of the laser to both ion and electron kinetic energies. In contrast to the few tens of eV temperatures typically produced in laser irradiation of monatomic gases, cluster targets can produce electron energy distributions in the few keV range, mean ion temperatures of 10–50 keV and peak ion kinetic energies up to 1 MeV. In addition, charge states up to 40^+ can be produced with quite modest laser intensities ($\approx 10^{16}$ Wcm^{-2}). Cluster targets are also surprisingly efficient (>90%) at absorbing intense laser light, and thus provide a new route to producing very high energy density, highly ionized laboratory plasmas of interest to a broad range of disciplines. Here we review recent experimental results and outline some areas of current research in this new field.

1. Introduction

The interaction of short pulse lasers with both solid and gas targets at intensities up to 10^{18} Wcm^{-2} has received considerable attention in recent years as it is of great interest to a number of diverse fields. These include basic plasma physics, laser fusion, high flux pulsed ion sources, high harmonic generation and the construction of bright X-ray sources for lithographic applications. There are very significant differences between the interaction physics seen in solid and gas targets, and the plasma conditions produced in these two media are correspondingly quite distinct. Solid targets irradiated with high intensity ($> 10^{16}$ Wcm^{-2}) sub-picosecond pulses produce near solid density plasmas with peak electron temperatures of order 1 keV, and exhibit absorption efficiencies of ≈50%. In contrast monatomic gas targets are typically very inefficient (<1%) at absorbing intense laser light, and quite cool (10–50 eV) plasmas produced by multi-photon and field ionization processes dominate. The fact that gas and solid targets interact in such different ways with intense lasers has prompted an investigation of the transition regime between the two, and this has proven to be a very fruitful area of research.

There are a number of targets that might prove suitable to fill the gap between solids and gases. Working down from the size regime of macroscopic solids, these include low density foams [1], structures such as diffraction gratings or metal blacks with micron scale modulations [2] and sprays of micro-droplets [3]. Moving up in size from monatomic gases, we might also consider investigating increasingly large molecules such as aromatic ring structures and the Fullerenes [4,5]. Atomic clusters occupy an important position in this hierarchy, as they can be produced in a broad range of sizes from a few to well in excess of 10^6 atoms/cluster. The interaction of atomic clusters with intense lasers has received considerable attention in the last few years [6–12] because, while some aspects of their behavior in intense fields are similar to those of solids or gases, they also show several distinctly new and quite unique effects. In particular gases of atomic clusters with a few thousand atoms/cluster are very efficient (>90%) at absorbing intense laser light [7], and produce extremely hot ion plasmas with mean temperatures of order 10–50 keV and peak ion kinetic energies approaching 1 MeV [9,10]. Ion charge states up to 40^+ are also seen in high Z clusters, at laser intensities 4 orders of magnitude smaller than would be required for a direct field ionization process alone in an isolated atom. In this review we summarize the basics of the laser/cluster interaction, report on several recent developments in the field and outline some possible directions for future work.

2. Cluster production in gas jets

Expansion of gas through a nozzle into vacuum results in substantial cooling in the frame of the moving gas, and atoms or molecules that interact weakly at room temperature can form clusters as a result [see Ref. 13 for a good general review]. Attractive forces between atoms due to van der Waals or hydrogen bonds become important, and the clustering process is primarily determined by the initial temperature and pressure of the gas reservoir, geometry of the nozzle, and strength of the inter-atomic bonds formed [14,15]. As with many complex fluid dynamics problems, there is no rigorous theory to predict cluster formation in a free-jet expansion. However, the onset of clustering, and size of clusters produced can be usefully described by an empirical scaling parameter Γ^* known as the Hagena parameter [15,16]:

$$\Gamma^* = k\frac{(d/\tan\alpha)^{0.85}}{T_o^{2.29}}P_0 \tag{1}$$

where d is the nozzle diameter (mm), α the expansion half-angle ($\alpha = 45^\circ$ for sonic nozzles, $\alpha < 45^\circ$ for supersonic) P_o the backing pressure (mbar), T_o the pre-expansion temperature (kelvin) and k a constant related to ease of bond formation.

Clustering typically occurs for $\Gamma^* > 100-200$, with the number of atoms per cluster scaling as $N_c \propto \Gamma^{*2.0-2.5}$ [14,17]. The strong variation of N_c with T_o and P_o can thus be used to engineer a medium of arbitrary mean density and cluster size. Mean atomic densities of 4×10^{19} atoms/cc, and cluster sizes from a few to $> 10^4$ atoms are easily achievable with quite simple pulsed jet designs [18] using a broad range of light and heavy gases. Various optical scattering techniques can be used to estimate mean cluster size in a jet expansion [19], but it is worth noting that a detailed characterization of such a jet (particularly the cluster size distribution) is experimentally very challenging.

2. A time-scale for the cluster expansion

Atomic clusters are typically small compared to the wavelength of light and break up rapidly when average ion kinetic energies exceed a few eV. We can make a simple estimate of the time-scale τ for the destruction of an atomic cluster in a strong laser field by considering the thermally driven expansion of a small collection of atoms from near solid density n_o to the average density n_a of the gas as a whole.

$$\tau \approx r_o (m_i / Z k_b T_e)^{1/2} (n_o / n_a)^{1/3} \tag{2}$$

where r_o is the initial cluster radius, m_i the ion mass and T_e the electron temperature. A 10^4 atom Ar cluster abruptly ionized to Ar^{8+} would have an initial radius $r_o \approx 50$ Å and assuming $n_a \approx 10^{18}$ atoms/cc, $n_o \approx 2 \times 10^{22}$ atoms/cc and $k_b T_e \approx 1$ keV gives $\tau \approx 1$ picosecond. We should thus require short pulse lasers if we wish to carry out interaction experiments with a cluster medium rather than a weakly ionized monatomic gas. In practice this condition may be somewhat modified in dense cluster media as there can be significant attenuation and steepening of the leading edge of a longer pulse as it propagates through a cluster medium [7].

3. Single cluster interaction physics.

A laser focused directly under the nozzle of a pulsed gas jet might interact with $> 10^{14}$ clusters on a single shot, and so to investigate single cluster effects as distinct from bulk cluster target physics, skimmers and long flight tubes are used to reduce the mean cluster density to a point where cluster-cluster interactions are negligible. Figure 1 shows such an experimental arrangement employed to investigate electron and ion kinetic energy distributions. Clusters from a free-jet expansion pass through a skimmer and the resulting low density cluster beam is intersected by a 150 fs, linearly polarized laser pulse at 780 nm from a Ti:Sapphire laser focused to an intensity of up to 2×10^{16} Wcm^{-2}. Ion energies are determined by time of flight to an MCP detector, electron energies by use of a hold-off grid, and ion charge states by a triple grid potential barrier. Figure 2 shows an electron energy distribution from 2500 atom Xe clusters. The important features are the very high kinetic energies seen [8] compared to a monatomic gas target (2.5 keV versus 10–50 eV) and the dual peak structure. Rotating the direction of the laser polarization vector, it is found that the "hot" electron peak at 2.5 keV is essentially isotropic, but the "warm" electrons at 800 eV are peaked along the laser electric field vector. Investigating the ion kinetic energies through time of flight (Figure 3) shows that mean energies of 45 keV and peak energies of 1 MeV are also observed. Again, the kinetic energies expected from the coulomb explosion of a small molecule

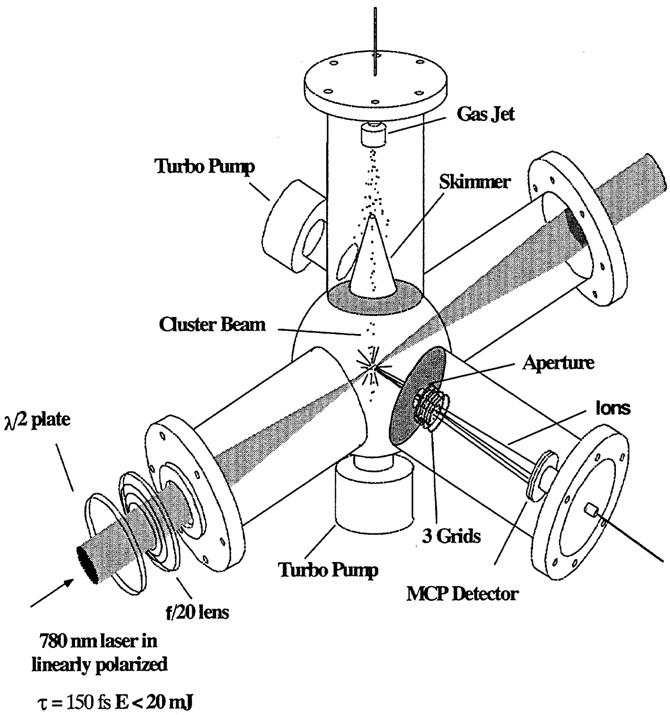

Fig. 1. Experimental arrangement for making electron and ion kinetic energy measurements in single cluster interaction experiments.

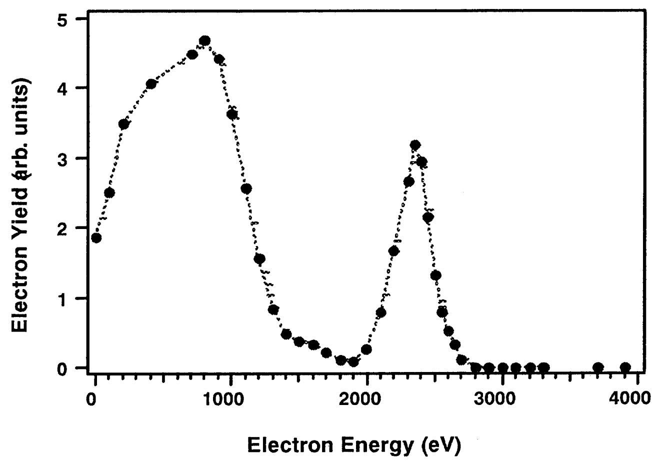

Fig. 2. Electron energy spectrum from the interaction of 2500 atom Xe clusters with a 150 fs, 780nm laser pulse focused to an intensity of 2×10^{16} Wcm^{-2} showing a distinct two component distribution.

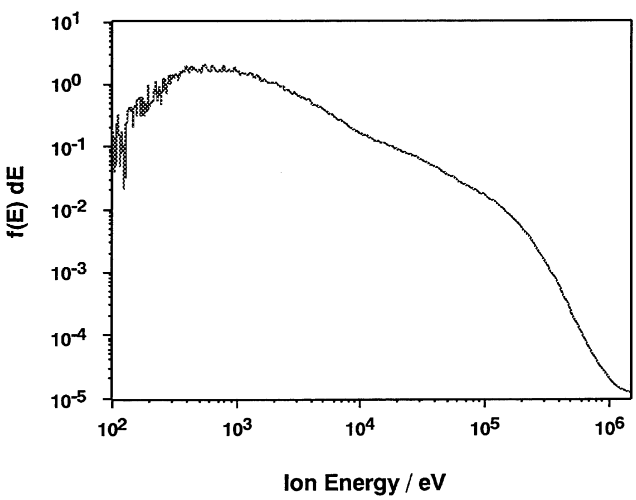

Fig. 3. Ion kinetic energy distribution from the interaction of 2500 atom Xe clusters with a 150 fs, 780 nm laser pulse focused to an intensity of 2×10^{16} Wcm^{-2} showing peak energies of ≈ 1 MeV.

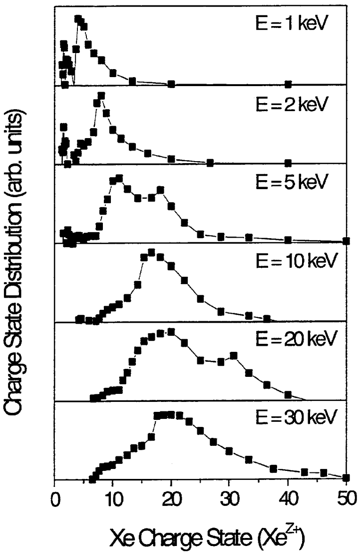

Fig. 4. Charge state distribution as a function of kinetic energy for 2500 atom Xe clusters irradiated with a 150 fs, 780nm laser pulse focused to an intensity of 2×10^{16} WCm^{-2}.

in a laser field of this intensity are of order 10–100 eV. Finally, examining the charge state distribution shows that ions up to Xe 40^+ are seen, with higher charge states typically having higher kinetic energies (Figure 4). To produce this degree of ionization directly with a laser field would require an intensity of order 10^{20} Wcm^2. Effects of this type can be seen with clusters of just a few hundred atoms, and ion kinetic energies from the cluster explosion are typically larger for higher Z materials, larger clusters and shorter wavelength heating pulses.

4. A simple model for the laser/cluster interaction

It is clear that the interaction of even relativly small clusters with an intense laser pulse is significantly different from that of a monatomic gas. Several models have been proposed to explain the anomalously high electron and ion energies and charge states observed in experiments. These include a resonantly driven coherent electron wave within the cluster [6] that preferentially produces inner shell ionization, and rapid collisional heating followed by an energetic explosion driven by a combination of hydrodynamic and Coulomb pressure, in which the Coulomb component plays a greater [20] or lesser role [21,22]. This last model has been particularly successful in explaining many experimental observations.

We can identify several distinct stages of the laser/cluster interaction. At the leading edge of the pulse a small number of free electrons are produced by multiphoton or field ionization processes. As the cluster is small compared to the wavelength of light, the electric field of the laser is to first order uniform across the cluster, and free electrons within it are driven by the laser electric field. As the density within the cluster is initially near solid, the collisionality is high, and electron impacts efficiently transfer energy to atoms within the cluster. The cluster begins to heat up, additional ionization occurs, and the resulting microplasma starts expanding. Early in the expansion the electron density n_e rises above 3 times critical ($n_{crit} = m_e\varepsilon_o\omega^2/e^2$ with m_e the electron mass, ε_o the permitivity of free space, ω the laser angular frequency and e the electron charge) and the atoms within the cluster are shielded from the laser electric field. As the cluster expands further (and for a suitable duration laser pulse the external electric field intensity rises) n_e falls and the cluster again comes into resonance with the laser field when $n_e = 3n_{crit}$.

At this point there is an electric field enhancement as the field E inside the cluster is a function of the external field E_o and the dielectric properties of the cluster microplasma: $E = 3E_o/|\varepsilon + 2|$ where the dielectric constant $\varepsilon = 1 - \omega_p^2/\omega(\omega + i\nu)$ for ω_p the plasma frequency and ν the electron/ion collision frequency. Electrons within the cluster are strongly driven, many electron/ion collisions occur producing high charge states, and hot electrons with energies of order 2–3 keV begin to escape from the cluster. The cluster then undergoes a very energetic explosion driven by a combination of Coulomb repulsion between highly charged ions, and hydrodynamic pressure as hot electrons stream away from the microplasma. There is however a degree of uncertainty as to which mechanism dominates in some circumstances.

5. Control of cluster dynamics

The existence of a resonance in the cluster heating at $n_e = 3n_{crit}$ that depends on both laser wavelength and plasma density allows us to control the cluster explosion dynamics to some degree. We can use a low intensity short pulse to start the cluster expansion, followed by an intense heating pulse that arrives when conditions for coupling laser energy into the cluster microplasma are optimum [23]. As the critical density depends on the laser wavelength, we can also use several different color heating pulses and hit a resonance at several points in the cluster expansion [24]. In both cases the peak ion energies achievable can be significantly enhanced.

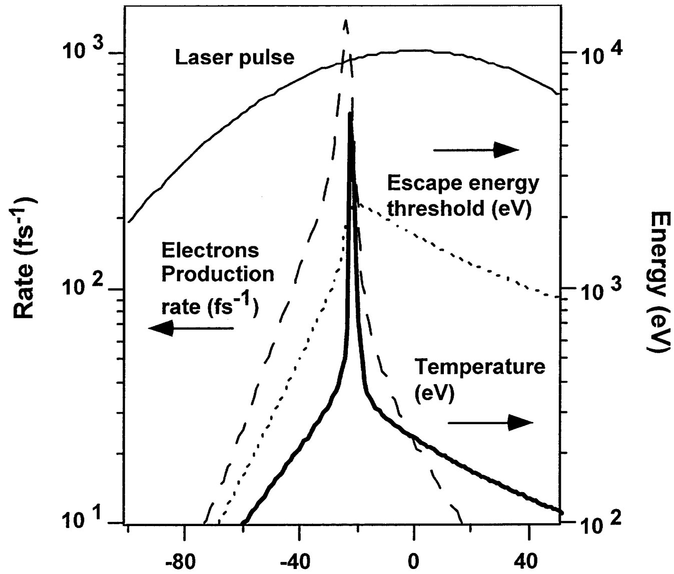

Fig. 5. Numerical modeling of the interaction of a 65 Å diameter, 2500 atom Xe cluster with a 150 fs 780 nm laser pulse focused to an intensity of 2×10^{16} Wcm^{-2}, based on an expanding spherical microplasma. The electron production rate (- - -), electron temperature (—) and escape energy threshold (. . .) inside the cluster are shown as a function of time.

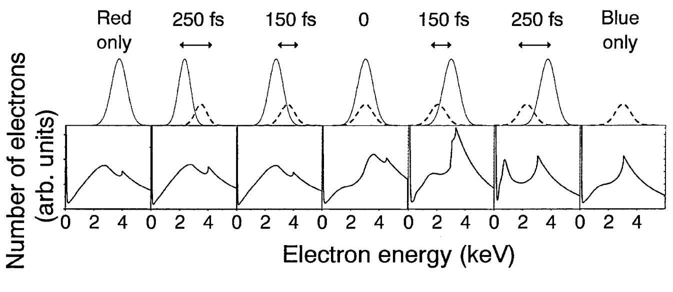

Fig. 6. Numerical modeling of 2500 Å Xe cluster interacting with 780 nm (solid line) and 390 nm (dashed line) laser pulses at 2×10^{16} WCm^{-2} for a range of time delays showing strong enhancement of the electron temperature for the 390 nm (blue) pulse 150 fs before the 780 nm (red) pulse.

Figure 6 shows the electron energy distribution produced by modeling the interaction of 150 fs, 780 and 390 nm laser pulses with 2500 atom Xe clusters as a function of the delay between the two heating pulses. There is a significant enhancement of the electron energy with the 390 nm pulse set 150 fs before the arrival of the 780 nm pulse, and in experiments this is found to produce an enhancement of $\approx 50\%$ in the mean ion energy observed [24].

6. Bulk cluster targets

Gas jets can easily be produced with average atomic densities of $\approx 5 \times 10^{19}$ atoms/cc [25] and thus laser interaction experiments carried out close to a jet nozzle typically involve $> 10^{14}$ clusters. In this high density regime, both cluster-cluster interactions in the expansion phase of the hot microplasmas and laser propagation effects caused by significant attenuation of the leading edge of the heating pulse become important. Due to the large number of clusters in the laser focus, absorption of laser energy into the bulk medium can be extremely efficient, approaching 100% [7]. Kinetic energy from cluster explosions is also thermalised as adjacent cluster microplasmas merge, and fast electrons from the cluster explosions heat the surrounding cold cluster gas [26]. Significant yields of X-rays in the keV range can be produced [27] and these may be of use for short wavelength lithographic applications due to the lack of target debris. In addition, these plasmas allow investigations of extremely high energy density plasmas at near solid density to be carried out with modest experimental facilities. Whilst the time scale for the cluster explosion is extremely short, spectroscopic measurements have shown that strong X-ray emission from high density plasmas are possible [28] indicating that there is significant X-ray emission from the clusters, whilst they are still at near solid density. This provides an interesting route to conducting experiments with high density plasmas, without some of the opacity problems associated with solid target experiments. Figure 7 for example shows strong X-ray emission from an Xe cluster plasma in the 12-15 Å range, with 4f-3d transitions in Xe^{26+} from satellite lines associated with a near solid density.

The fragility of the clusters also provides an interesting route to engineering useful electron density or temperature profiles in a cluster medium. A low level prepulse can be used to break up clusters in localized regions of the jet leaving a cold monatomic gas. Following this by an intense heating pulse results in the regions still containing clusters becoming

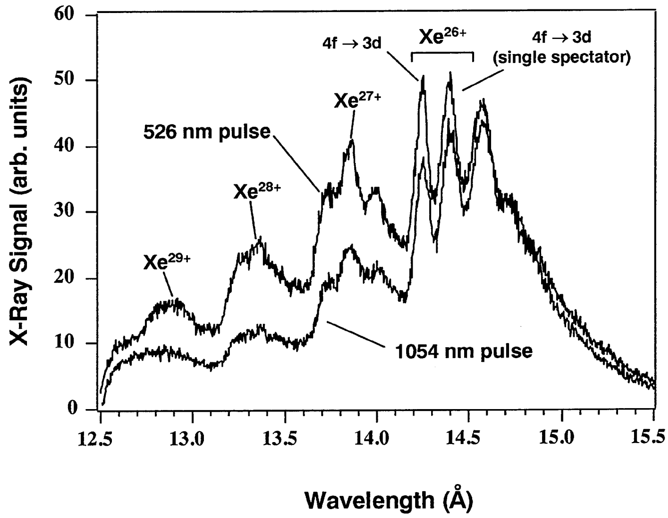

Fig. 7. Time integrated X-ray spectra from a Xe cluster plasma when 2.5 ps, 526 nm and 1054 nm pulses are used to heat the clustering gas, showing strong 4f-3d transitions in Xe^{26+} from satellite lines.

extremely hot and highly ionized, whilst the cluster-free regions remain relatively cold. This can be used for example to produce annular electron density profiles for guiding intense laser pulses through plasmas [29].

7. Microdroplet Targets

A new extension of the cluster interaction field is the investigation of micro-droplet targets in which a large number of ≈ 1 μm diameter liquid droplets are irradiated by an intense laser. These droplet sprays can be produced by expelling liquid under pressure through a small nozzle [3] and can in principle be doped with a wide range of materials. This would greatly extend the range of elements that can be used in interaction experiments beyond that of clusters.

There are several reasons to expect droplet targets to produce interesting effects. The droplets are large enough that skin depth effects and the non-uniform laser electric field across the droplet become important. They may be too large to exhibit a strong dielectric resonance as seen in clusters, but they are sufficiently large that a plasma density gradient can be set up around each droplet allowing efficient coupling by classic resonance absorption that can transfer laser energy into extremely hot electrons, and inertial compression of the core of the droplet occurs as the outer layer ablates away. However, unlike solid targets in which there is rapid cooling by electron thermal or radiative conduction to the cold bulk of the target, the heated droplets should also remain hot until hydrodynamic cooling takes place. Preliminary investigations of droplet targets show them to be very efficient at absorbing intense laser light, with coupling fractions similar to solid targets. However, due to the geometry of the droplets, the absorption is invariant with polarization, and unlike solid targets does not appear to decrease at very high ($< 10^{17}$ Wcm^{-2}) intensities. As with cluster targets, the droplet spray is extremely efficient at converting laser energy into X-rays, however the ease with which droplet sprays can be doped with a wide range of elements suggest that they may be easier to optimize for applications requiring a bright source in a particular wavelength range.

8. Conclusions

The interaction of intense, short pulse lasers with atomic clusters has been an extremely fruitful area of research in the last few years and has brought to light several new and interesting pieces of physics. The laser-cluster interaction can be extremely energetic, and produces plasmas considerably hotter than would be obtained with either solid or monatomic gas targets irradiated at a similar intensity. To date, mean ion energies of 40–50 keV and peak ion energies of 1 MeV have been demonstrated using table-top scale short pulse laser systems and it seems likely that optimization of the cluster size and heating pulse duration, or multi-pulse heating schemes can be used to enhance this significantly. The cluster interaction can be understood in terms of a dynamic resonance that enhances the electric field inside the cluster as it interacts with a short, intense laser pulse, followed by an energetic explosion. This allows us to modify the cluster explosion dynamics through the use of multiple heating pulses.

The great efficiency of energy coupling into cluster media means that they are interesting both as sources of high energy density plasmas for research in areas such as astrophysics, and as bright X-ray sources for lithographic and other applications. It is unclear whether cluster sources will also be useful as a source of highly charged ions for seeding accelerators, where solid target laser plasmas are quite well developed. However, they do provide a very interesting route to conducting a broad range of high energy density experiments with modest experimental facilities, and can routinely generate very large concentrations of high charge state ions on extremely short timescales.

Acknowledgements

The majority of the work reported on in this review was carried out in the Blackett Laboratory Laser Consortium at Imperial College, London and we gratefully acknowledge the support of the EPSRC, MOD and BNFL, and the technical assistance of P. Ruthven, A. Gregory and S. Hanif. R. Smith is supported by an EPSRC Fellowship.

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