

# Femtosecond Pulse Physics

## The intensity of contrast enhanced Astra Gemini laser



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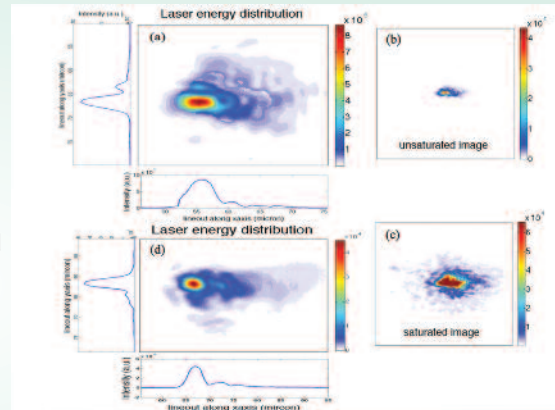
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The intensity in the laser focus is one of the key issues in the high-intensity laser-solid interactions. Astra Gemini laser delivers more than 4 J in a single pulse with FWHM duration of 50 fs, allowing for laser intensities in excess of  $5.6 \times 10^{20} \text{ W/cm}^2$ . Here we present a complete analysis on the measured laser focal spot image in a recent campaign employing nm-thin foils and calculate the actual laser intensity with experimental conditions. This analysis may explain why the ion energies obtained in this campaign were below 20 MeV, which at a first glance is puzzling by virtue of the large amount of laser energy delivered to the target. Compared to other laser systems, it also shows the need for adaptive optics to improve on this issue.



(a) Laser energy distribution in logarithmic scale. (b) Measured (blue) and fitted (red) curve for the readout per pixel. (c) Table of different relevant intensity values for Astra Gemini laser system. With the same conditions for a perfect Gaussian beam, the encircled energy ratio is 0.5.

## A two-screen spectrometer to measure the 3D momentum distribution of GeV electron beams produced by laser wakefield acceleration experiments on Astra Gemini



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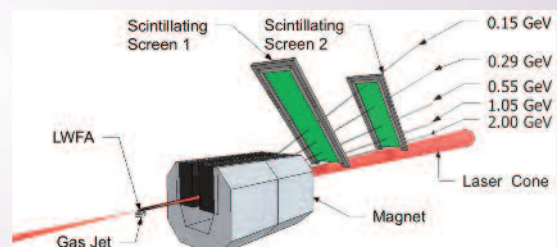
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Laser wakefield accelerators (LWFAs) have great potential for future high-energy physics applications, as compact sources of high-energy particles, and as light sources. This is in part due to their ability to sustain accelerating fields ( $> 100 \text{ GV/m}$ ) around three orders of magnitude greater than can be supported in conventional radio frequency accelerators ( $< 100 \text{ MV/m}$ ).



Experimental setup. Electrons of differing energies are shown passing through scintillating screens as calculated by tracking algorithm.

## Near-GeV electron energy acceleration in low density plasma channels



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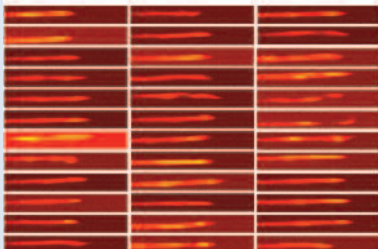
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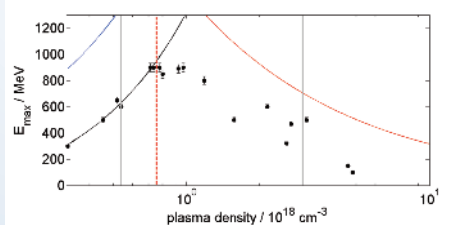
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The Astra-Gemini laser was used to study electron acceleration driven in plasma channels. Electrons with energies up to 900 MeV were generated and the properties of these beams studied with respect to the laser and plasma parameters. The variation of the electron energy with the density of the plasma was found to agree well with a simple model of the acceleration process. The shot-to-shot reproducibility of the generated electron beams – which is important for

applications of laser-accelerated electron beams, such as radiation generation – was also studied. Under optimum conditions, electron beams were generated every laser shot; the energy spectrum of the electrons was found to be relatively stable, with most of the variations being attributable to variations in the energy and pointing of the driving laser.



Electron spectra from 36 consecutive shots are shown. For each image the horizontal axis is increasing energy from left to right.



The maximum recorded electron energy as a function of the axial plasma density. The density for which the dephasing length equals the capillary length is shown by the dotted red line. Expected electron energies according to theory are shown in red and blue. A fit to the data (black) shows good agreement with the expected linear dependence of electron energy on plasma density.

## A two-screen electron spectrometer for broad- and narrow-bandwidth electron spectra



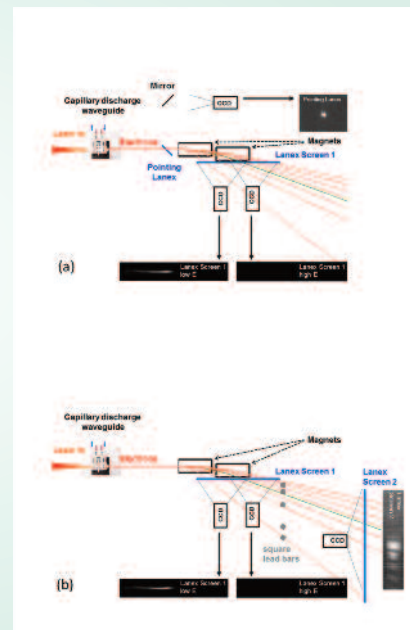
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Magnetic dipole spectrometers are widely used to determine the energy of electron beams by recording their deflection in a known magnetic field. Accurate energy measurement requires that the trajectory of the beam as it enters the magnetic field is known, but in practice the pointing of beams generated by laser-driven accelerators varies with each laser shot. We have studied two methods for measuring the incident electron beam trajectory without disturbing the measurement of its energy spectrum. For the conditions of our experiment, these methods were shown to lead to energy corrections of approximately 10%.



Sketch of the experimental set-up for the Pointing Lanex (a) and the two-screen (b) methods of energy correction.

## Electron acceleration up to 2 GeV in plasma channels

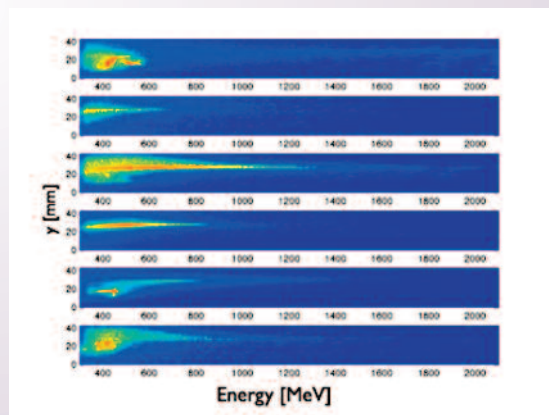


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Laser wakefield accelerators develop as compact sources of short-pulse relativistic electrons. A relativistic structure with close to gigavolt per centimetre fields is created in the laser wake on a plasma background. These structures capture electrons and increase their energy along the region where the laser is kept at high-intensity. The accelerator efficiency is expected to improve by propagating the laser in a preformed plasma channel acting in the laser as a fiber optic.

We have used 4 centimetre long plasma channels and the Astra-Gemini laser in a configuration close to external guiding resulting in the acceleration of electron up to 2 GeV. We have also used a gas cell to produce mono-energetic electron bunches of 500 MeV. Therefore, we have demonstrated the two basic blocks of a double-stage electron accelerator for acceleration of mono-energetic electron bunches beyond 2 GeV.



Energy spectra of electrons produced by LWFA in a plasma channel with maximum energies up to 2 GeV in shots 1 and 3.

## Calibration of grazing-incidence flat-field soft x-ray diffraction gratings



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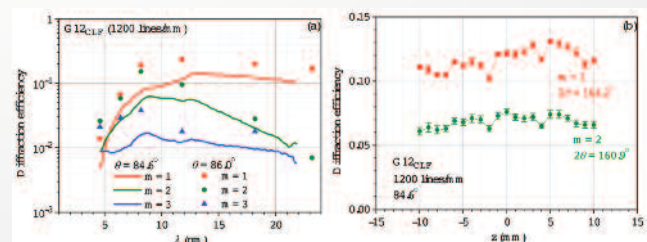
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Grating-based x-ray spectrographs are widely used in experiments. Among the factors contributing to the spectrograph throughput, the grating efficiency is typically the least known factor and is subject to change with time. We report on measurements of the diffraction efficiency of three gratings used in the recent experiments on relativistic high-order harmonics generation in gas jet targets with the Astra Gemini laser in CLF, RAL, UK and the J-KAREN laser in KPSI, JAEA, Japan. The diffraction efficiencies at the incidence angles coinciding with those in the

experiment were measured using a reflectometer installed at a soft x-ray beamline of SR Center, Ritsumeikan University, Japan. The CLF grating having two narrow strips on the surface caused by laser irradiation shows the high uniformity of efficiency parallel to the groove directions except shallow dips at the strip positions. The reported results are indispensable for accurate spectral measurements and quantitative data analysis.

Lines: diffraction efficiencies for the spectral orders  $m = 1, 2,$  and  $3$  of  $G12_{CLF}$  at  $\theta = 84.6^\circ$  measured at BL-11. Dots: earlier measurements at  $\theta = 86.0^\circ$ . (left graph).

Position dependence of diffraction efficiencies for the spectral orders  $m = 1$  and  $2$  measured parallel to the groove directions of  $G12_{CLF}$ , at fixed  $\lambda = 10.0$  nm,  $\theta = 84.6^\circ$ ,  $2\theta = 164.2^\circ$  for  $m = 1$  and  $2\theta = 160.9^\circ$  for  $m = 2$ .



# Non-linear relativistic plasma optics in GeV laser wakefield accelerator

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Self-guiding of a relativistically intense 55 fs laser pulses over 15 mm leading to temporal compression down to 10-20 fs has been observed. Measurements of the transmitted pulse energy, as the gas density is varied, reveal the pump depletion process during the interaction and how this relates to the acceleration of high-quality GeV electron beams. Estimates of the

dephasing length indicate that, in the case of the most mono-energetic beams, the pulse is modified and compressed for approximately half of the interaction before the acceleration begins. Power amplification of the input beam of up to 40% was seen for densities where no significant electron beam was generated.

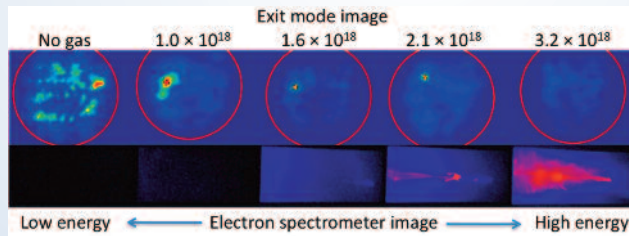


Figure 1: (Top) Spatial distribution of the transmitted laser pulse showing the vacuum beam size (red circle) and (Bottom) spectrometer screens showing the accelerated electrons (high energy to the right). The electron densities given above each pair of images are in  $\text{cm}^{-3}$ .

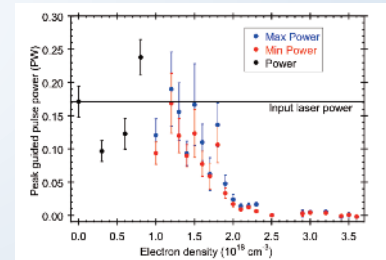


Figure 2: The peak pulse power plotted against gas electron density. The minimum and maximum powers are due to the uncertainty in the pulse duration.

# High Energy Laser Interactions

## Super critical electrostatic shocks in laboratory plasma



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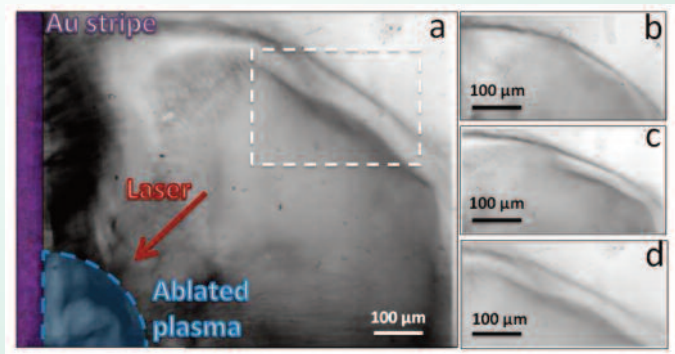
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Collisionless shocks are relevant to a variety of astrophysical scenarios, and have recently been the focus of several laboratory plasma investigations employing high-power lasers. A study of the generation and evolution of collisionless shocks was carried out using the Vulcan laser. The shocks are generated during the expansion of a warm laser-produced plasma into a tenuous ( $\sim 10^{16} \text{ cm}^{-3}$ ), non-magnetized background plasma. Shock structures form at the edge of the expanding plasma, and are detected via

proton radiography. In particular, we have identified in the data the transition from a unipolar field profile, typical of a double layer structure, into a bipolar electric field, and the subsequent formation of collisionless, electrostatic shocks, which propagate in the ambient plasma with a Mach number  $\sim 4$ . A PIC simulation supports the existence of high Mach number ( $\sim 3.3$ ) shocks launched by collision of plasma clouds of equal electron and ion temperatures.

(a) Proton Image of the interaction of nanosecond pulse with  $50\mu\text{m}$  Au stripe, showing shock structure created during the expansion of laser-ablated plasma into tenuous plasma.



## A comparison of different radiochromic film types



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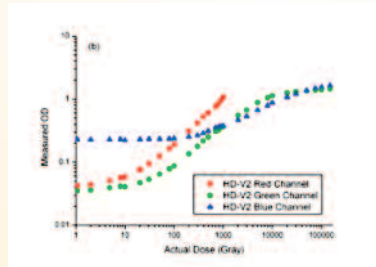
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Radiochromic film (RCF) is a self-developing, passive dosimetry film consisting of an active layer which when exposed to radiation undergoes a polymerization reaction, increasing its optical density in proportion to deposited dose.

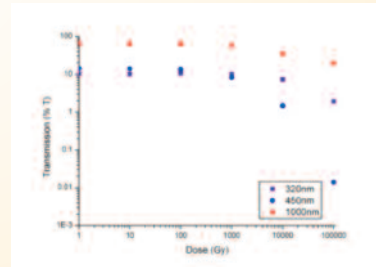
This report presents work carried out to produce absolute calibration curves for various forms of radiochromic film. It discusses the now discontinued HD-810 and presents a comparison with its replacement, HD-V2. Results show that

HD-V2 is less sensitive than its predecessor, but can potentially give a greater dynamic dose range. A third, more sensitive form of radiochromic film, called EBT-2 is also characterized and discussed, including its significant potential to be used in conjunction with HD-V2 to provide 'novel' stack designs.

Preliminary work is also presented on extending the dose range for HD-V2 and EBT-2 using infra-red or ultraviolet wavelengths.



The relationship between true optical density and dose for each of the RGB channels, scanning HD-V2.



The transmission of ultraviolet, infra-red and blue wavelengths through HD-V2 at various, known pre-exposed doses.

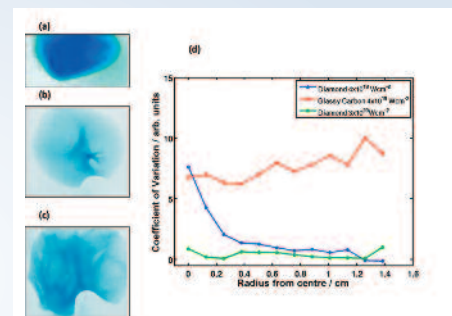
## The effect of lattice structure on fast electron transport in warm dense carbon



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In this report, we present preliminary results from an investigation of the effect of lattice structure on fast electron transport. Using various forms (allotropes) of the same element, carbon, we show that lattice structure can determine whether or not fast electron beam filamentation occurs in solid targets irradiated by intense picosecond laser pulses. By investigating this at two laser intensities,  $4 \times 10^{19} \text{ Wcm}^{-2}$  and  $3 \times 10^{20} \text{ Wcm}^{-2}$ , we also show that the onset of electron beam filamentation is sensitive to the drive laser pulse energy. These findings may have a direct implication in the choice of targets for laser-driven ion sources and advanced schemes of inertial confinement fusion.



Spatial-intensity profile measurements of the proton beam, at energies of 10 MeV for (a) diamond at  $3 \times 10^{20} \text{ Wcm}^{-2}$  and 5.6 MeV for (b) diamond at  $4 \times 10^{19} \text{ Wcm}^{-2}$  and (c) vitreous carbon at  $4 \times 10^{19} \text{ Wcm}^{-2}$ . (d) Radial variance analysis of the proton signal for each target material, in which the variation in the proton beam dose as a function of radius is calculated.

## X-ray scattering from liquid carbon heated by laser-accelerated protons

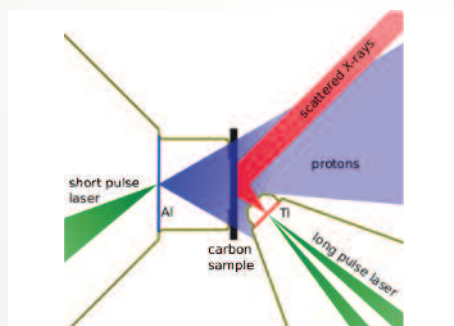


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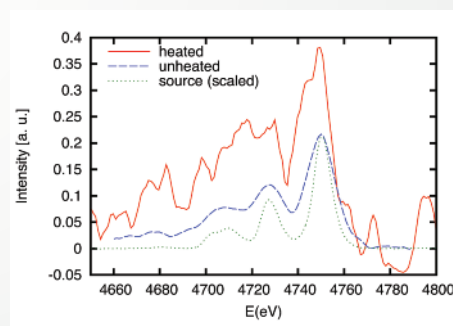
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We used laser-accelerated protons to partly melt carbon samples and observed the phase transition using X-ray scattering. Carbon melt data can serve as a test for warm dense matter theories, but is also relevant in astrophysics, as liquid carbon may exist in the core of giant planets. Vulcan's short pulse beam 8 was focused on a thin aluminium foil to accelerate protons from the rear side which then heated a graphite sample to temperatures

of a few thousand Kelvin. Due to the material's inertia hydrodynamic expansion was negligible and isochoric heating was achieved. X-rays from a laser-generated titanium plasma were scattered from the sample at 90° scattering angle. At this scattering angle, the structure factor of carbon increases strongly when the sample melts. The transition is therefore clearly visible in the scattered spectrum.



Target.



Measured x-ray spectra. The source spectrum is scaled to size to illustrate the spectral form of the source.

## Modelling channel formation by high intensity laser plasmas

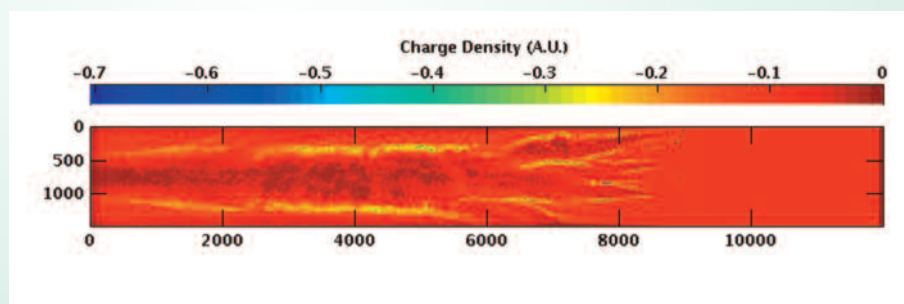


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This report represents results from 2D PIC simulations with the code OSIRIS showing plasma channels created by focusing either a 1ps or 30 ps pulse with peak intensity of  $3 \times 10^{18} \text{ Wcm}^{-2}$  into a hydrogen gas. This channel is produced (primarily) from the radial expulsion of plasma due to the ponderomotive force of the laser. The size of the plasma channel was measured to be

mm in scale and the dynamics of the process involve laser self-focusing and filamentation of the plasma channel for higher densities. A good match is found in most cases between the simulations and previously reported experimental observations, though small deviations occur at higher densities.



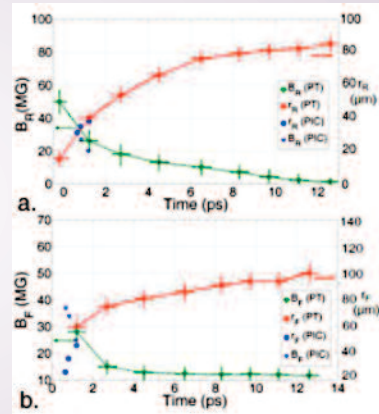
## Dynamics of self-generated, large amplitude magnetic fields following high-intensity laser matter interaction



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We report here on simultaneous measurements of the magnetic fields generated at the front and at the rear side of a solid target irradiated by a short and intense laser pulse, using a spatially and temporally resolved proton imaging technique. Data analysis gives evidence of the generation of toroidal magnetic fields (BMAX ~ 50 MegaGauss) that decay in time on a picosecond time scale. Their spatial distribution and amplitude is consistent with the recirculation of the laser-accelerated electrons around the target and it is seen to confine the radial expansion of the plasma. Beside a direct connection to ion acceleration scenarios, these plasma conditions have some relevance to astrophysical scenarios, e.g. leptonic jets.



Experimental measurements of the temporal evolution of the magnetic field amplitude and width at the rear (a.) and front (b.) side of the target compared with PIC simulations.

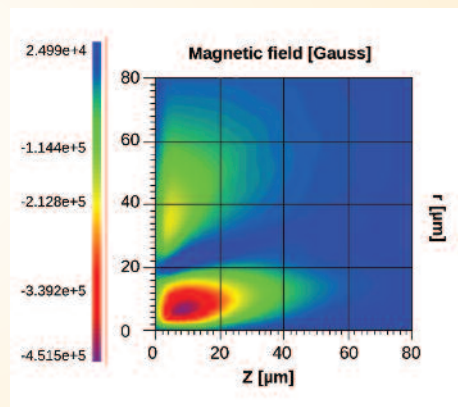
## Controlling fast electron beam divergence using two laser pulses



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This paper describes the first experimental demonstration of the guiding of a relativistic electron beam in a solid target using two colinear, relativistically intense, picosecond laser pulses. The first pulse creates a magnetic field that guides the higher-current fast-electron beam generated by the second pulse. The effects of intensity ratio, delay, total energy and intrinsic prepulse are examined. Thermal and K $\alpha$  imaging show reduced emission size, increased peak emission and increased total emission at delays of 4 - 6 ps, an intensity ratio of 10:1 (second:first) and a total energy of 186 J. In comparison to a single, high-contrast shot, the inferred fast-electron divergence is reduced by 2.7 times, while the fast-electron current density is increased by a factor of 1.8. The enhancements are reproduced with modeling and are shown to be due to the self-generation of magnetic fields. Such a scheme could be of considerable benefit to fast ignition inertial fusion.



The modelled magnetic field at the time when the second pulse is beginning for the case where the guiding was optimal ( $t_{delay} = 4.7$  ps). The negative field near the axis collimates the fast-electrons.



## X-ray scattering from warm dense iron



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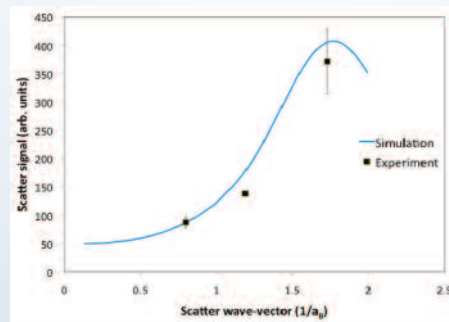
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Warm dense iron is of great interest from the perspective of planetary physics and geophysics. Understanding the microscopic structure would help make great advances in understanding the thermodynamics and mechanics of planetary structure and formation. To this end, X-ray scattering from warm dense matter can make a significant contribution.

We have created warm dense samples of Fe by laser-drive shock heating and compression using the Vulcan laser.

The shock speed has been evaluated by optical streak pyrometry and it has been established that the Fe is expected to be in a molten state at several 100s GPa pressure.

X-ray scattering from the sample has been recorded at three angles simultaneously and over multiple shots up to five angles have been recorded for a given sample condition. We will present our data, initial analysis and conclusions.



*Comparison of experimental results with HNC simulation. Conditions for iron in simulation are  $T_e = 2$  eV,  $\rho = 10$  g/cc and  $Z^* = 6$ .*

# Theory and Computation

## Multi-electron and multi-channel effects on harmonic generation



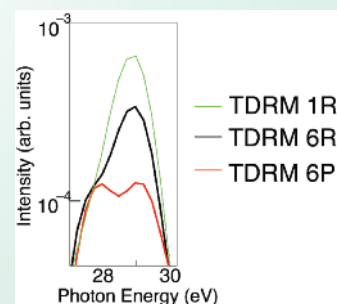
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Harmonic generation is a fundamental process occurring when intense laser light interacts with matter. By studying this process, we can obtain fundamental insight into the movement of electrons in atoms and molecules. A key question is how the dynamics of different electrons is linked together. As a means of investigating this, we have developed time-dependent R-matrix theory and its extension to study harmonic generation. We have applied the approach to He atoms interacting with 390nm laser light to study the accuracy of different possible schemes to calculate harmonic spectra.

*By systematically improving the basis set for the description of, in this case, He, we can assess the effect of atomic structure on harmonic spectra. The most simple basis shows a structureless peak for the 9th harmonic, but a double-peak structure becomes noticeable in the most sophisticated one.*

We have also applied our method to investigate how autoionising resonances affect the magnitude of the fifth harmonic of Ar in the 200-240nm range. The appearance of these resonances indicates strong interference between the response of different Ar electrons to the laser light. Hence, harmonic generation demonstrates that the dynamics of different Ar electrons is strongly connected.



## A new equation of state for waterbag-distributed plasma



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In previous work we deduced an upper bound on the amplitude of the electric field in a class of 3D waterbag solutions to the Vlasov-Maxwell system. Unlike for 1D waterbag solutions, it was shown that the electric field is finite as the phase velocity of the wave tends to the speed of light. In that calculation the Vlasov equation was

tackled directly, without recourse to the stress-energy-momentum tensor. In order to shed further light on the behaviour of the plasma, the present article focusses on a derivation of the relationship between the stress-energy-momentum tensor and proper number density of the electron fluid described by the 3D waterbag.



## Development and validation of a 1D2V Vlasov-Fokker-Planck model

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Inertial fusion energy (IFE) has the potential to provide a clean, safe, abundant, secure and complementary method of generating useful electrical energy. However, developing an efficient IFE scheme is reliant on effectively coupling a radiation source e.g. a laser to an IFE fuel capsule. Coupling occurs via electrons and thus a detailed understanding of their transport behaviour under the extreme conditions

generated by inertial confinement fusion is required. A 1d2v Vlasov-Fokker-Planck model capable of simulating this behaviour has been developed and is nearing the end of an extensive validation phase that has yielded promising results. The construction of the model and the results of the suite of validation tests form the focus of this paper.

## Approximating the dynamic structure factor in warm dense matter



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The memory function model was used to examine the behaviour of the dynamic structure factor in a one-component plasma. The results were compared to

those from MD simulations, and further restrictions on the behaviour of static structure factor models obtained.

## The effect of temporal pulse shape in simulating the interaction of an intense laser pulse and an ultra-thin foil

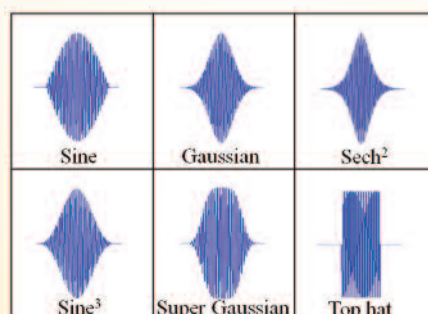


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Ultra-intense laser-plasma interactions can be used to establish accelerating quasi-electrostatic field gradients on the rear surface of a solid target that are orders of magnitude higher than the current limit of conventional, radio-frequency-based accelerator technology. However a detailed understanding of how these bright proton beams can be controlled and optimised is required in order to harness their application potential.

With optical control methods in mind, 1D Particle-In-Cell simulations of an intense laser interaction with an ultra-thin foil have been carried out to investigate the effect of varying the shape of the temporal envelope of the pulse on the maximum proton energy of the subsequent proton beam.



The six different pulse shapes employed in the simulations described in this paper.

## Radiation reaction in ultra-intense laser fields



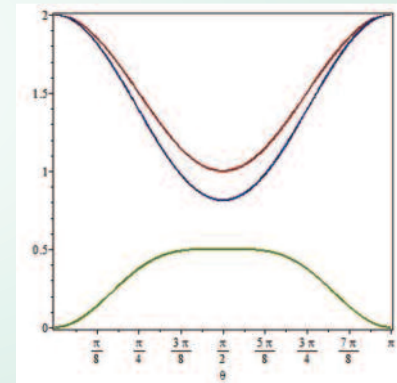
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We discuss various corrections to standard Thomson scattering between an electron and a laser, in particular radiation reaction, nonlinear and quantum effects. For each of these we identify a characteristic dimensionless parameter and determine the leading order correction term. We point out that a unified description of the

different mechanisms has been achieved in terms of strong-field QED – with the exception of radiation reaction. Nevertheless, our discussion suggests that radiation reaction effects might become enhanced through large nonlinearities while quantum effects still remain small.

Angular dependence of the differential cross sections for Thomson scattering in units of  $r_e^2 \pi$ .  $r_e$  is the classical electron radius and  $\theta$  the scattering angle. Dimensionless laser amplitude  $a_0 = 0.5$ . Red: standard Thomson scattering. Blue: fundamental harmonic for nonlinear Thomson scattering. Green: Second harmonic correction.



## The study of beam-plasma instabilities relevant to laser-plasma interactions in fast ignition

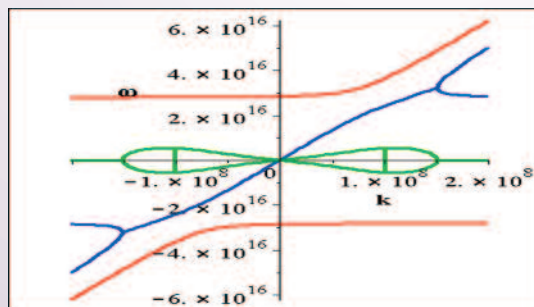


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We present results from numerical simulations conducted to investigate a potential method for realizing the required fusion fuel heating in the Fast Ignition scheme to achieving inertial confinement fusion. The heating mechanism investigated is the relaxation of the non-thermal electron beam through the two-stream instability. This process generates Langmuir waves that parametrically decay into lower amplitude Langmuir waves and ion acoustic waves that are strongly

damped by ion collisions in the dense plasma, resulting in energy transfer to the background plasma ion population. The results demonstrate energy transfer to the ion population from the laser driven electron beam via the non-linear wave-wave interaction associated with the two-stream instability. Evidence will also be provided for the effects of competing instabilities such as the Weibel and filamentation modes found to be detrimental to the ion heating process.



Two-stream dispersion solution for a beam-plasma system with background plasma density of  $10^{29} m^{-3}$ , beam density of  $10^{28} m^{-3}$  and beam energy of 1MeV. The red line is the real root for the plasma contribution to the dispersion, the blue line is the real component from the beam and the green line is the imaginary component of the solution from the beam.

## QED-PIC Codes for 10PW laser-plasma simulation

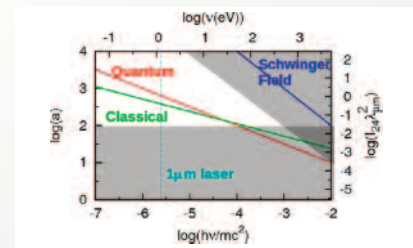


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With construction beginning soon on several next-generation 10PW lasers as part of the European Union's Extreme Light Infrastructure(ELI) project, an exciting new frontier will soon be reached in highpower laser-plasma physics. 10PW lasers will create strong enough electromagnetic fields to access non-linear quantum electrodynamics (QED) processes. It will therefore be essential to include such processes in the modelling of laser-plasma interactions as laser powers reach and surpass this point. We propose a simple model for doing this – a QED-PIC code. This model relies on two basic assumptions, that the laser fields can be treated as: (1) quasi-static; (2) weak. The first assumption relies on the strength parameter of the laser  $a > 100$ , the second relies on the peak laser electric field being less than 1% of the Schwinger field for vacuum breakdown. The figure opposite shows where these assumptions are met (the

unshaded region). This corresponds to the most interesting regime for next generation laser plasma interactions, taking in both the region above the green line where classical radiation reaction is important and above the red line where the process becomes quantum mechanical. This broad validity of QED-PIC means that this approach will provide a very powerful simulation tool for future high intensity laser matter interactions.



Domain of validity for QED-PIC.

## ZEPHYROS v.0.7 series



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The CLF's PPG has made its 3D particle hybrid code ZEPHYROS available to users. Here we describe how we have modified

the code and continued to improve it to both fully meet user requests and expectations and accelerate productivity.

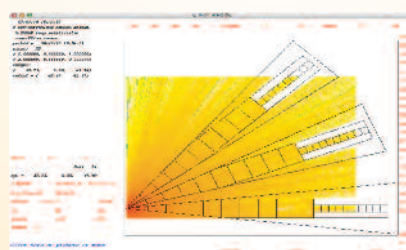
## The spectral distribution and total kinetic energy of fast electrons generated in a relativistically intense, frequency doubled laser-solid interaction



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Novel bremsstrahlung photon detectors have been successfully fielded on a relativistically-intense laser-solid experiment using frequency doubled laser light. Analysis of the experimental data was performed with the 3D Monte Carlo code MCNPX. The spectral distribution of the bremsstrahlung photon spectrum generated



by the fast electron beam corresponds to a relativistic Maxwellian of temperature  $125 \pm 25$  keV, this is best fit by Sherlock's scaling law. The measured laser to fast electron conversion efficiency was  $13 \pm 3\%$  at an intensity of  $5 \pm 3 \times 10^{18} \text{ Wcm}^{-2}$ . The design of the bremsstrahlung photon detector is described and the interpretation of the frequency doubled experimental data using Monte Carlo modelling is detailed.

*The modelled spatial distribution of bremsstrahlung ux of all energies. The target is at the bottom left of the image, while the lead shielding of the three detectors is visible as the white regions (low photon ux) towards the top and right.*

## Developing an integrated approach to modelling short pulse laser-solid interaction



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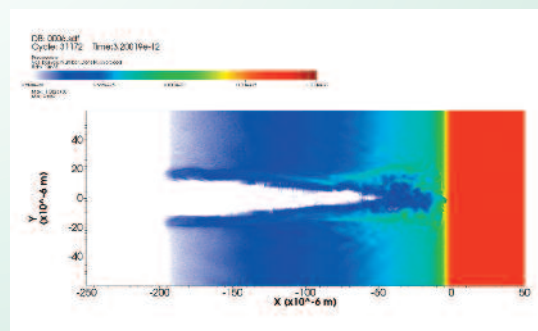
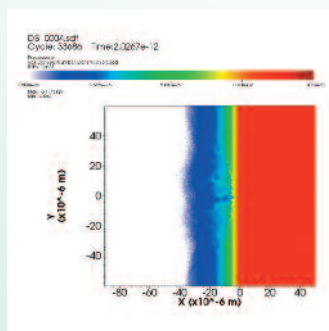
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Modern Particle in Cell (PIC) codes, Vlasov-Fokker-Planck models and radiation hydrodynamic codes together constitute the tools required to model key aspects of short pulse laser-plasma interaction. However a complete description of the problem requires a daunting breadth of physics, which cannot reasonably be included in a single code. Our approach is to develop an integrated modelling capability in which detailed models for different aspects of the problem are linked together.

We outline our methodology and demonstrate the approach taken to link PIC models of LPI into hybrid models of electron transport for a number of different target conditions – focussing on the impact of changes in pre-plasma scale length and target material on target heating.

*Electron density profiles from EPOCH for 50µm (top) and 10µm (bottom) scale-length pre-plasmas. In the long scale length plasma, a stable channel is formed. In both cases, filamentation as the beam approaches critical densities generates an unstable hot electron source which changes direction as the plasma evolves.*



## Wave breaking and saturation of resonantly driven waves in warm plasma



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A unified theory for the growth and breaking or saturation of driven waves in warm, inhomogeneous plasma is presented. This theory incorporates both secular growth and convective saturation of the waves, and incorporates earlier results for growing waves in cold plasma

[Koch and Albritton, Phys. Rev. Lett. 32, 1420 (1974)] and saturated waves in warm plasma [V.L. Ginzburg (Gordon and Breach, New York, 1961)]. It also discusses previous attempts to reconcile these two limiting cases.

## Monte Carlo studies of ion-ion inverse Bremsstrahlung absorption



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A Monte Carlo code has been developed to study ion-ion inverse Bremsstrahlung absorption in the presence of a linearly polarised laser field in high intensity regimes of  $10^{20}$  W/cm<sup>2</sup> and above. It has been benchmarked against several test problems, including the calculation of

transport coefficients (see fig. 1).

The ion-ion inverse Bremsstrahlung heating mechanism allows for direct absorption of laser energy by ions, and is shown in fig. 2 to generate heating in the keV in a 0D model with a suitable choice of ion species and a 30fs pulse length at critical density.

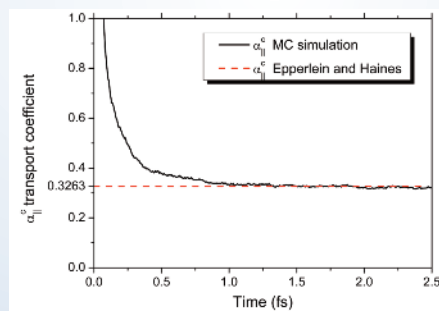


Figure 1.

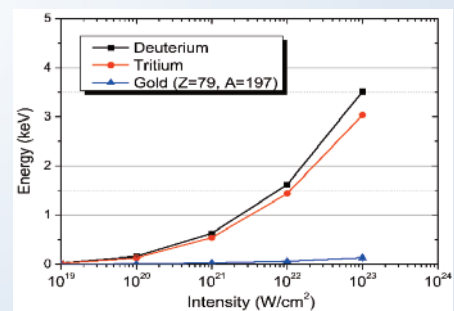


Figure 2.

# Ultrafast and XUV Science

## Ultrafast pump-probe experiments with an exemplar biomolecule, phenylalanine



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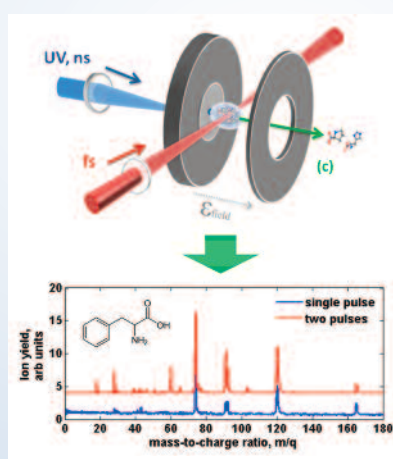
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A biomolecular gas phase target is created by using the LIAD technique (top), whereby a UV, ns laser irradiates the reverse side of a foil, launching a shock wave to liberate molecules deposited on the front. The plume is crossed by a fs laser, and resulting ions are extracted to a time of flight device. The recorded mass spectrum (bottom) can uncover signatures of ultrafast processes, such as charge transfer.

For the first time, an ultrafast pump-probe study with a complex molecule has been performed.

Biomolecules were liberated into the gas phase via laser induced acoustic desorption (LIAD), creating a target plume of intact neutrals. In this pump-probe scheme, a



Mach-Zehnder interferometer was used to generate two fs pulses, delayed from each other by  $\tau_D$ . The first pulse acts to ionise the neutral biomolecules, and the second to probe the remaining ions. The resulting ions, generated at a series of delays, are then extracted to a time of flight mass spectrometer. Studying changes in fragmentation with delay times should allow ultrafast charge migration processes to be resolved.

Although this study has not provided evidence of ultrafast charge transfer in phenylalanine, there are a number of possible explanations. Future studies will further investigate these, and this experiment has shown the viability and promise of such a scheme in examining ultrafast processes.

## Towards UV pump XUV probe photoelectron spectroscopy of chemical dynamics



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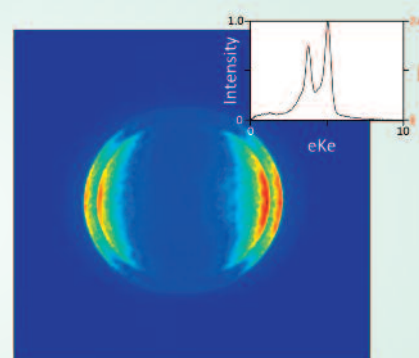
S. Weber, M. Siano and J.P. Marangos  
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Preliminary experiments aimed at the development of UV pump XUV probe photoelectron spectroscopy at the Artemis laser facility have been performed. The experiments involved the monochromated high harmonic generation beamline combined with the velocity map imaging spectrometer.



Photoelectron image of Xe taken with the 11th harmonic. Inset: retrieved photoelectron spectrum obtained via the POP algorithm



## Coherent collective-mode oscillations in the $K_{0.3}MoO_3$ charge density wave



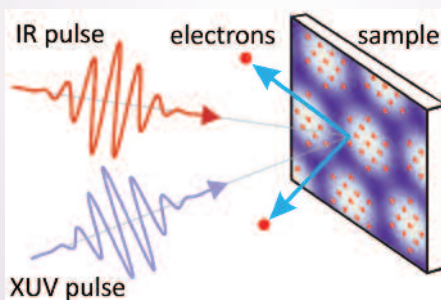
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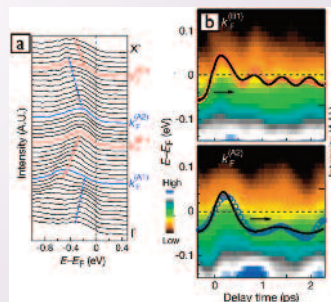
The total energy of a low-dimensional metal can be lowered by forming a charge density wave (CDW), distorting the crystal lattice and often producing a metal-to-insulator transition by opening a gap in electronic states at the Fermi energy. New collective modes also appear, corresponding to oscillations of the amplitude and phase of the CDW. To understand how these collective modes couple to the band structure and to each other, we performed time- and angle-resolved photoemission spectroscopy at Artemis in the CLF. We measured the

momentum-dependent electronic structure of blue bronze ( $K_{0.3}MoO_3$ , a linear chain compound), evolving in time after optical excitation.

The different CDW gaps melt, recover, and then oscillate: one at the amplitude-mode frequency, the other at the phase-mode frequency. This first observation of coherent phase mode dynamics, which are Raman-inactive, is uniquely permitted by our photoemission probe. The phase excitation may reflect anharmonicity in the strongly-driven order parameter.



*In time-resolved ARPES, an ultrashort laser pulse creates a transient phase in a material. A subsequent XUV pulse generates photoelectrons. Collected and analysed, these reveal the momentum- and energy-dependent electronic structure as it evolves on femtosecond time scales.*



*(a) Momentum-dependent photoemission spectra of blue bronze. (b) Response to ultrafast stimulation, measured at the Fermi wave vectors. Line-outs show integrated intensity near EF. One gap oscillates at the amplitude-mode frequency and one at the phase-mode frequency.*

## The commissioning of the AMO end station at Artemis: first steps toward laser induced electron diffractions



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When an atom or a molecule is subject to a strong laser field the potential barrier which keeps electrons bound to the parent ion is weakened. If the field intensity is high enough the tunnel effect can occur (tunneling ionization) in which an electron is promoted into the continuum from the highest occupied molecular orbital. For

electrons released into the continuum just after the peak of the electric field and for linearly polarized light it can be shown with very simple classical arguments that the electrons return on the parent ion around half a field cycle later. Consequently recollision with the parent ion can occur.

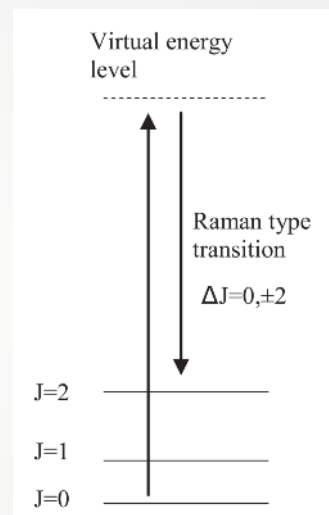
## Exploring the onset of superfluid behaviour in quantum clusters using time-domain measurements



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Many features of superfluidity can be broadly described in terms of the absence of friction. Here, CO molecules were attached to small helium clusters to probe molecular superfluidity on the nanoscale. Our aim is to investigate how many helium atoms are needed to produce evidence of superfluidity and how slow or fast rotating molecules respond to a superfluid environment. To deliver this ambitious aim, an entirely new method to obtain this information is applied: we excite rotational wavepackets of single molecules in superfluid helium and study how these wavepackets propagate in time. This will directly show how the rotational speed is affected by a potential superfluid environment and it will also show how local superfluids, in close proximity to a molecule, respond to instantaneous rotational excitation.



*Schematic showing the sequential excitation and de-excitation of rotational levels in Raman cycles.*