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Exotic states of matter explored with short-wavelength free-electron lasers

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Outline



(a) interaction of intense short-wavelength radiation with matter: sources of motivation; volumetric heating (b) short-wavelength lasers used for these studies (i) plasma-based lasers (ii) free-electron lasers (c) short-wavelength laser ablation and desorption (d) characterizing the focused beam from its imprints (e) behaviour of matter in short-wavelength laser microfocus (i) soft X-rays (92 eV): FLASH + probing plasma (ii) x-rays (1.56-1.83 keV): LCLS (f) summary

Why interaction of short-wavelength laser radiation with solids should be investigated?



 (1) Estimating and minimizing damages to surfaces of high-flux irradiated XUV/X-ray optical elements developed and used for the guiding and focusing of short-wavelength laser beams as well as those used for long-term irradiation with high repetition rate sources,
 (2) durability assessments of materials suggested for the first walls of ICF reactors and optical elements exposed to intense X-ray radiation in a laser-plasma interaction chamber,

(3) diffraction-limited ultrastructuring and patterning of solid surfaces for fabrication of microelectronic and micromechanical elements and devices,

(4) determination of radiation field characteristics: imaging of spatial energy distribution in a focused beam ablatively imprinted on the irradiated material and determination of pulse energy content, and (5) production of very dense plasmas with $T_e \sim 10 \text{ eV}$ (WDM - warm dense matter) and $T_e \sim 100 \text{ eV}$ (HDM - hot dense matter).

Warm Dense Matter - WDM

Nonideal character of plasma is usually characterized by the coupling parameter mentioned by S. Pascarelli last Thursday.

$$\Gamma = \frac{U}{E_k} \simeq 1$$







volumetric heating

In certain position, where the frequency of electrons oscillating in plasma (plasma frequency; Langmuir

frequency) is equal to the frequency of the laser EM field, the index of refraction of the plasma becomes zero. Thus the EM wave cannot penetrate into the plasma, being reflected. This is called plasma mirror effect.

The plasma frequency is a function of the plasma electron density

$$\omega_{\rm e}^2 = n_{\rm e} \, {\rm e}^2 \, / \, \varepsilon_0 \, {\rm m}_{\rm e}$$

and the laser frequency is equal to the plasma frequency exactly at the critical electron density

 $n_{\rm c}$ [electrons/cm³] = 10²¹ × λ^{-2} [µm]

for λ < 10 nm is n_c > 10²⁵ cm⁻³

 \Rightarrow X-rays do not create a critically dense plasma so that their energy is deposited in a volume below the irradiated solid surface





Casser des molécules, une des éventuelles applications du laser X. (p. 16)



P. Jaeglé: Le laser à rayons X, *La Recherche* (184), 16-25 (Jan. 1987).

...decomposition of molecules, one of possible applications of X-ray lasers

Neon-like zinc XRL driven by multi-100-ps IR laser pulses

Simplified level scheme of neon-like zinc



NANOSCOPY LAB

Generic experimental scheme



Active medium: a plasma column created from slab target by linearly focused IR laser beam







Composite linear focusing optics with 30-mm clear aperture



A novel arrangement: matrix of 10 cylindrical lenses 150x60 mm + single f[#]3 aspherical lens





Smaller cylindrical segments may be fabricated very precisely for a moderate cost !



highly uniform & laterally tight linear focus

XRL driving conditions: plasma keV self-emission





Prepulse: $\sim 1.6 \times 10^{10} \text{ Wcm}^{-2}$

single cylindrical + spherical lens deliberately de-focused

Main pulse: ~2.8×10¹³ Wcm⁻² composite optics: high-quality line focus

Cross-slit X-ray camera data IP-ASCR-Prague



Summary of parameters of the Ne-like Zn laser



Wavelength \Leftrightarrow hv	21.22 nm ⇔ 58.53 eV
XRL pulse energy	4 mJ
XRL pulse length	80-100 ps
Peak power	40 MW
Photons per pulse	3×10 ¹⁴
Brightness* ($\Delta\lambda/\lambda=0.001$)	10 ²⁷ phot s ⁻¹ mm ⁻² mrad ⁻²
No. of shots with one "batch"	~100

* Brightness (brilliance) = $\frac{\text{No. of "monochromatic" photons } (\Delta \lambda / \lambda = 10^{-3})}{\text{sec } \times \text{ surface unit } \times \text{ solid angle unit}}$

focusing scheme of Ne-like Zn soft x-ray laser







table-top capillary-discharge XUV laser

[made in Fort Collins, Colorado State University – S. Heinbuch et al.: *Opt. Express* **13**, 4050 (2006)]



46.9 nm

0.01 mJ

1-2 ns

<u>10 Hz</u>

Capacitors

Capillary

Laser output



installed in Prague

Focusing the CDL beam

- spherical Sc/Si multilayer mirror (f = 0.25 m) with maximum reflectivity at 46.9 nm
- angle between the incident and reflected beams was about 6°
- sample holder with the target was placed in 3D positioning stage
- irradiated samples of PMMA poly(methyl methacrylate) were illuminated and observed with CCD camera equipped with a magnifying objective





• 15 shots for were accumulated



FLASH - Free-electron LASer in Hamburg





TESLA Test Facility (TTF 1 FEL, 1995-2002)

FLASH, 2005

experimental hall

Photon energy	~30-300 eV
Bandwidth $\Delta\lambda/\lambda$	~0.5 %
Peak power	>1 GW
Pulse duration	~10-100 fs

The European X-Ray Free-Electron Laser

Technical design report

1 Introduction

This Technical Design Report of the European X-Ray Free-Electron Laser (XFEL) Facility has been prepared by a large community of scientists and engineers and was edited at Deutsches Elektronen-Synchrotron (DESY) laboratory under the supervision of the European XFEL Project Team.

J. Krzywinski a kol.:

Conductors, semiconductors and insulators irradiated with short-wavelength freeelectron laser,

J. Appl. Phys. <u>101</u>, 043107 (2007)



X-Ray Free-Electron Laser

Figure 1.2.1 Interaction of powerful Vacuum Ultraviolet (VUV) radiation with solids [1-1]. Ablation of Gold target after one pulse of the VUV Self-Amplified Spontaneous Emission (SASE) FEL at the TESLA Test Facility (TTF) at DESY. Radiation wavelength is 98 nm, pulse duration is 40 fs, peak power density is about 100 TW/cm².

References

[1-1] L. Juha et al., Ablation of various materials with Intense XUV radiation, Nucl. Instrum. and Methods A 507 (2003) 577.

FLASH experimental hall



K. Tiedtke et al.: New J. Phys. 11, 023029 (2009)





Ablation of monocrystalline silicon by a train of ten pulses of 32-nm FEL radiation (BL2 at FLASH, Fall 2005).

Three regimes can be distinguished in PMMA response to intense soft x-ray radiation



DESORPTION REGIME – below threshold process of single photon material removal occurring at beam tales and creating shallow craters (depth ~ 1nm)

INTERMEDIATE REGIME – steep rising edge of the efficiency curve. Unstable regime involving both: ablation and desorption.

ABLATION REGIME – above threshold process of collective material removal creating deep craters (depth ~ attenuation length)

$$\eta(\varepsilon) = \frac{n_R(\varepsilon)}{n}$$

Here, the material removal efficiency in a single-compound material is a ratio between dose dependent density of removed atoms and total density of atoms in unirradiated material. Red curve in the figure has a meaning of zero approach to real efficiency curve.

Single-shot damage to PMMA by focused FLASH beam



A shallow crater created by a pulse of radiation from FLASH at 21.7nm. The fluence is slightly above the threshold (~10mJ/cm²) distinguishing the ablation, intermediate, and desorption regimes of material removal (A – the ablation region, I – the intermediate region, D – the desorption region).

A crater created by the focused beam of FLASH at 21.7nm distinguishing between ablation and desorption regimes. Blue area represents mild surface modification caused by desorption.





AFM surface roughness measurement for particular interaction regimes.



This is not a MD simulation!

Desorption of PMMA

Few soft x-ray photons cannot decompose the PMMA main chain efficiently, but the probability of fragment release is not zero. Desorption of the material starts, occurring at low fluences.



my

Ablation of PMMA

Many soft x-ray photons decompose more efficiently the PMMA main chain. The probability of fragment release is higher and starts saturating. Ablation of the material occurs.

May My



m

Desorption\ablation model and crater morphology

Let's assume the local absorbed dose in the form:

$$\mathcal{E}(x, y, z) = \mathcal{E}_0 f(x, y) e^{-z/l_{at}}$$

 Satisfying the condition of local and sudden response, i.e., neglecting any longitudinal and transverse energy transfer from irradiated region to the pristine material (charge carrier diffusion, heat conduction), we are allowed to express the crater morphology as follows:

$$d(x, y) = l_{at} \int_{0}^{\varepsilon_0 f(x, y)} \frac{\eta(t)}{t} dt$$

• Where:

- •I_{at} ... PMMA attenuation length
- $\bullet \epsilon_0 \dots$ local peak dose absorbed in the material
- •f(x,y) ... transverse intensity profile
- •η(ε) ... dose dependent material removal efficiency

Dose-dependent crater profile simulation



A perfect Gaussian beam profile is assumed in the simulation. Parameters of the material removal efficiency curve were taken from the measurement $(\eta_D \sim 0.07, \eta_A = 1, \epsilon_D = 1460 \text{ a.u.}, \epsilon_A = 1580 \text{ a.u.}).$

Method of material removal efficiency measurement

• Assuming a perfect Gaussian probe beam $\varepsilon(x) = \varepsilon_0 \cdot \exp(-x^2/\rho^2)$, the material removal efficiency curve is to be determined from a transverse crater cross-section measured by means of AFM as follows:

$$\eta(\varepsilon) = -\frac{\rho^2}{2xl_{at}} \cdot d'(x) \bigg|_{x=\pm\rho\sqrt{\ln(\varepsilon_0/\varepsilon)}}$$

- Where:
 - x ... transverse coordinate
 - d'(x) ... first derivative of the transverse crater profile cross-section
 - ρ ... beam radius (at 1/e of maximum)
 - l_{at} ... attenuation length
 - $-\epsilon_0 \dots$ peak local dose





LAB

Experimental layout of the A/T measurement using microfocusing 13.7-nm FEL radiation by off-axis parabolic mirror (OAP)



Ablation imprints of the focused beam

- Transverse beam profile reconstruction
 - soft x-ray beam profile reconstruction utilizing ablative imprints in PMMA (poly(methyl metacrylate) to derive transverse irradiance distribution
 - the ablation imprint's morphology is investigated by means of the atomic force microscopy (AFM)

• Model assumptions and conditions:

- non-thermal ablation (no thermal melting)
- Lambert-Beer law of radiation absorption
- local energy deposition (no transverse heat or charge transfer)
- attenuation length << Rayleigh range
- irradiance-independent ablation threshold

$$f(x, y) = \frac{F_{th}}{F_0} \exp\left(\frac{d(x, y)}{l_{at}}\right)$$

Where:

- f(x,y) ... reconstructed beam profile d(x,y) ... measured crater profile F_{th} ... threshold fluence
- F_n... peak fluence
- I_{at} ... attenuation length





Z: 18.8a.u.

f(x,y)

X: 40.04m



J. Chalupsky et al.: Opt. Express 15, 6036 (2007).

- Advantages:
 - direct "in focus" detector (no propagation algorithms needed)
 - almost AFM-like resolution
- Disadvantages:
 - dynamic range limited by non-thermal and thermal ablation threshold
 - "ex situ" detector
 - limited wavelength range (surface RMS enhancing with the photon energy)
- Solution: high-Z materials (lead tungstate PbWO₄)

Reliable range







A.J. Nelson et al., Opt. Express 17, 18271 (2009)



An example of transverse beam profile reconstruction exploiting ablative imprints in PMMA. A sub-micron focus at 13.5nm has been achieved and characterized at FLASH.



Quiz question: What is the full width at half maximum (FWHM) of this beam profile?

Answer: FWHM is a bit misleading used for characterizing non-Gaussian beams.

Solution: Define the effective beam diameter.

F-scan technique

- fluence scan (F-scan) method = measurement of the beam cross-section area at various fluence levels
- derived from the beam profile reconstruction eq. for the surface contour:

$$f(x, y) = \frac{F_{th}}{F_0} \exp\left(\frac{d(x, y)}{l_{at}}\right) \xrightarrow{d(x, y)=0} f(x, y) = \frac{F_{th}}{F_0} = \frac{1}{p}$$

- the surface contour represents a beam contour at level 1/p of maximum
- varying the peak fluence F_0 , i.e. the peak-to-threshold fluence ratio p, we scan through the beam profile



An AFM scan (on the right) and Nomarski image (on the left) of an ablative imprint in PMMA created by 15-Å (830eV) LCLS radiation 26mm out of the focus (AMO station). Even though the surface roughness is increased, the surface contour remains preserved.



J. Chalupsky et al.: Opt. Express 18, 27836 (2010).



(i) advantages:

- the f-scan method enhances the dynamic and wavelength range of the ablative imprints methods by several orders of magnitude
- does not need AFM measurements
- very high resolution
- applicable to any material with an irradiance-independent ablation threshold
- good tool to characterize non-Gaussian beams
- (ii) difficulties:
 - needs a stable beam profile; shot-to-shot fluctuations must be minimized
 - not suitable for heavily attenuated beams (limited by the ablation threshold)
 - high-irradiance imprints may be influenced by piston effect and spallation

Why could be the high resolution of use?



- an ablative imprint in PbWO₄ created by 6Å LCLS radiation (2keV) out of the focus
- the pulse energy adjusted by a gas attenuator (T=1%)
- no Be filters used
- in principle usable for beam profile reconstruction at 2keV



- an ablative imprint in PbWO_4 created by 6Å LCLS radiation (2keV) out of the focus
- the pulse energy adjusted by a gas attenuator (T=2%)
- Be filters used (T=20%)
- a micron-sized speckle structure observed indicating the wavefront has been shattered by the Be filter

Effective beam area (A_{eff})

- is to be regarded as a reference quantity for all spot-size definitions (4σ , RMS, FWHM,...)
- provides mathematically correct relation between the pulse energy (E) and peak fluence (F_0)
- best approach to treat non-Gaussian beams
- measurable by means of the F-scan

Exercise \rightarrow define: $F(x, y) = F_0 f(x, y)$, where: $0 \le f(x, y) \le 1$ for $(x, y) \in R^2$ integrate: $E_{pulse} = F_0 \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} f(x, y) dx dy = F_0 A_{eff}$ effective beam area: $A_{eff} = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} f(x, y) dx dy$



The threshold-to-peak fluence ratio $f = 1/p = E_{th}/E_{pulse} = F_{th}/F_0$ as a function of the surface contour area S. PMMA and CVD diamond (chemical vapor deposition) samples were both irradiated at the same position by focused LCLS beam (830 eV, AMO station). The effective area is represented by the area below the measured curves.

Longitudinal beam profile characterization

• Stigmatic beams

- the shape of the ablative imprints an their dependence on the longitudinal z-position provide information about the focusing performance
- assuming a stigmatic Gaussian beam we may express the crater area (surface contour area) as:





Almost circular ablative imprints in PbWO₄ created by a stigmatic focused LCLS beam at 1600 eV (SXR station, perfectly aligned K-B optics, non-monochromatized "white" beam).

Measured crater areas in relation to the longitudinal zposition. Bars, circles, and triangles represent different attenuation levels. The data are fitted by the stigmatic Gaussian model function.



• Astigmatic beams

– an astigmatic beam is usually responsible for elliptical imprints



- the orientation of the major axis changes by 90 degrees when scanning through the focus
- from the fit of a theoretical Gaussian model we may judge about the astigmatism
- **assuming** an astigmatic Gaussian beam we may express the crater radii as:

$$r_{1}(z,F_{0}) = \rho_{1}(z) \left(\ln \frac{F_{0}}{F_{th}} - \ln \left(\frac{\rho_{1}(z)\rho_{2}(z)}{\rho_{01}\rho_{02}} \right) \right)^{1/2}, where: \rho_{1}(z) = \rho_{01} \left(1 + \frac{(z-z_{c1})^{2}}{z_{01}^{2}} \right)^{1/2}$$
$$r_{2}(z,F_{0}) = \rho_{2}(z) \left(\ln \frac{F_{0}}{F_{th}} - \ln \left(\frac{\rho_{1}(z)\rho_{2}(z)}{\rho_{01}\rho_{02}} \right) \right)^{1/2}, where: \rho_{2}(z) = \rho_{02} \left(1 + \frac{(z-z_{c2})^{2}}{z_{02}^{2}} \right)^{1/2}$$

Where:

 $\rho_{01},\,\rho_{02}\ldots$ beam waist radii

- $z_{c1}, z_{c2} \dots$ beam waist positions
- $z_{01}, z_{02} \dots$ Rayleigh ranges
- F_{th} ... threshold fluence

F₀ ... peak fluence

 $\rho_1(z), \rho_2(z) \dots$ beam radii at arbitrary z-position

Subscripts 1,2 denote sagittal and tangential plane, respectively.

Mutually coupled equations → must be fitted simultaneously.



Almost elliptical ablative imprints in PbWO₄ created by an astigmatic focused LCLS beam at 800 eV (SXR station, misaligned K-B optics, nonmonochromatized beam).

Measured horizontal and vertical crater diameters in relation to the longitudinal zposition. The data were fitted by the astigmatic Gaussian model functions in order to determine the astigmatic difference Δf , i.e., the distance between the horizontal and vertical focus.



$M_h^2 = (3.6 \pm 0.5)$ and $M_v^2 = (3.2 \pm 0.5)$

Challenges for future multi-shot desorption imprints in PMMA

- below-threshold material removal (desorption)
- multiple shots needed to modify the surface significantly
- the beam profile is proportional to the crater morphology



Multi-shot PMMA desorption z = 42mm imprints (LCLS/SXR station, monochromatized beam at <u>39µm</u> 800eV). Various numbers of z = 62mm shots 100,300, or 500 . accumulated. z = 82mm 23µm FOCUS 20µm z = 102mm z = 86 mm z = 122mm 16µm z = 142mm z = 104 mm 12µm z = 162mm 12µm z = 182mm z = 124 mm 12µm z = 202mm 18µm Single-shot PbWO₄ z = 144 mm z = 222mm ablative imprints 19µm (LCLS/SXR station, monochromatized beam at 800eV) 20 µm 26µm



Measured crater diameters (single-shots in PbWO₄) fitted by astigmatic Gaussian model functions (LCLS/SXR station, monochromatized beam at 800ev).

³⁰ - PbWO₄ - caustic curve (geom. avg.) • PMMA - effective radius ¹⁰ ¹⁰

An approximate caustic curve determined from the PbWO₄ measurements (geometrically averaged radii) compared to effective beam radii measured by means of multi-shot desorption imprints in PMMA. The results are in a good agreement.



LAB

experimental layout of the A/T measurement using microfocusing 13.7-nm FEL radiation by off-axis parabolic mirror (OAP)





Transmission of 52-nm Al foil (10-nm Al_2O_3) for the 13.7-nm FLASH beam focused by Si/Mo coated OAP at an irradiance ranging from 10¹⁴ W/cm² to 10¹⁷ W/cm² [B. Nagler a kol.: Turning solid aluminium transparent by intense soft X-ray photo-ionization, *Nature Physics* 5, 693 (2009)]

electronic structure of solid Al (L shell)





photo-ionizing one 2p electron from L shell





blue shift of the L edge; photo-ionization of more 2p electrons is not possible by 92-eV FEL photons









homogeneous heating is solving the problem of T_e and n_e gradients

PRL 104, 225001 (2010)

PHYSICAL REVIEW LETTERS

week ending 4 JUNE 2010

Electronic Structure of an XUV Photogenerated Solid-Density Aluminum Plasma

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an example of "a dilute system": low cross section and/or yield of the phenomenon investigated









OES of aluminium placed in the FLASH microfocus [10¹⁶ W/cm²; J. Cihelka et al.: Optical emission spectroscopy of various materials irradiated by soft x-ray free-electron laser, *Proc. SPIE* 7361, 73610P (2009)]

Computer simulation of AI I emission using the code MARIA





calculation: F. Rosmej, O. Renner



resulting excitation temperature is about 0.8 eV





OES of copper massive target placed in the FLASH microfocus [10¹⁶ W/cm²; J. Cihelka et al.: Optical emission spectroscopy of various materials irradiated by soft x-ray free-electron laser, *Proc. SPIE* 7361, 73610P (2009)]

Optical emission spectrum of Ce:YAG irradiated by 13.7-nm FLASH radiation focused at an irradiance of 10¹⁵ W/cm²







S. Toleikis et al.: one FALSH pulse heats and scatters (~8·10¹³ W/cm²)



LCLS:SXR experimental setup





S. M. Vinko et al.: Creation and diagnosis of solid-density hot-dense matter with an X-ray free-electron laser, *Nature* 482, 59 (2012)



FEL photon energy: 1830 eV



Conclusions

- First experimental results looking at high-intensity X-ray FEL interaction with solid density AI samples: dynamics are very different to single-atom case;
- Electron collisional processes dominate the CSD, even in the presence of the intense X-ray pulse;
 - simulations indicate 'thermal' CSD within a fraction of the pulse
- Temperatures in excess of 150-200 eV achieved at solid densities;
- Absorption/heating determined by the ionized states, ground-state cross sections largely irrelevant!
- Emission spectrum generated by a quasi-monochromatic X-ray pulse is not necessarily representative of the CSD, but is sensitive to the K-edges of the excited system.

S. M. Vinko et al.: Creation and diagnosis of solid-density hot-dense matter with an X-ray free-electron laser, *Nature* **482**, 59 (2012)





SACLA - <u>S</u>Pring-8 <u>Angstrom Compact free-</u> electron <u>LA</u>ser Commissioned: June 2011

Currently lasing at 0.08 nm

ELI-Beamlines Facility near Prague





ELI Beamlines Facility laser





ELI-Beamlines mission



- 1. Generation of femtosecond secondary sources of radiation and particles
 - XUV and X-ray sources (monochromatic and broadband)
 - Accelerated electrons (2 GeV 10 Hz rep-rate, 100 GeV low rep-rate), protons (200-400 MeV 10 Hz rep-rate, >3 GeV low-rep-rate)
 - Gamma-ray sources (broadband)

2. Programmatic applications of the femtosecond secondary sources

- Medical research including proton therapy
- Molecular, biomedical and material sciences
- Physics of dense plasmas, WDM, laboratory astrophysics
- 3. High-field physics experiments with focused intensities 10²³-10²⁴ Wcm⁻²
 - Exotic plasma physics (e.g. electron-positron pair plasma), non-linear QED
- 4. Participation in prototyping technologies for the high-intensity pillar Compression & coherent superposition of multi-10-PW ultrashort pulses

Summary



(1) Short-wavelength lasers interact with matter and plasmas in a different way than conventional, long-wavelength lasers.

- (2) Unique states of matter (e.g., WDM) can be both created and probed by short-wavelength free-electron lasers.
- (3) XUV/X-ray free-electron lasers produce from irradiated solids relatively unifom dense plasmas because of volumetric (caused by the short wavelength) and isochoric (resulting from delivering energy in ultra-short pulses) heating.



thank you for your attention