Phase transformations produced by intense fs-laser inside a crystal

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MOTIVATION

Generation of high pressure/temperature in sub-micron volume

Confined in a bulk of transparent and opaque solids

Imitating conditions in the cores of stars and planets

Formation of new super-dense phases

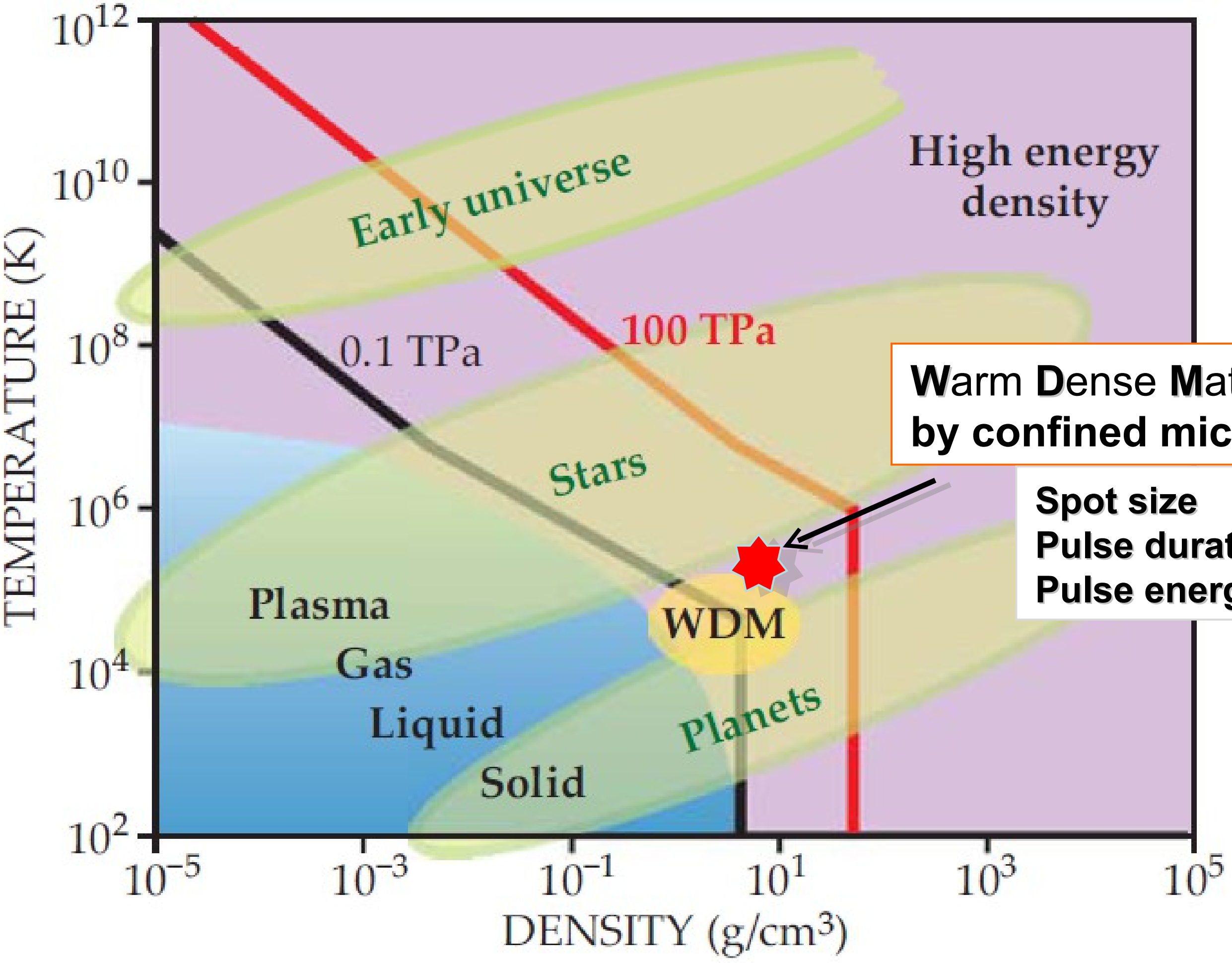
Observation, understanding and control of material behaviour

in laboratory table-top experiments

"To see a world in a grain of sand…" William Blake

High energy density states in the Universe

R. P. Drake, Physics Today, June 2010

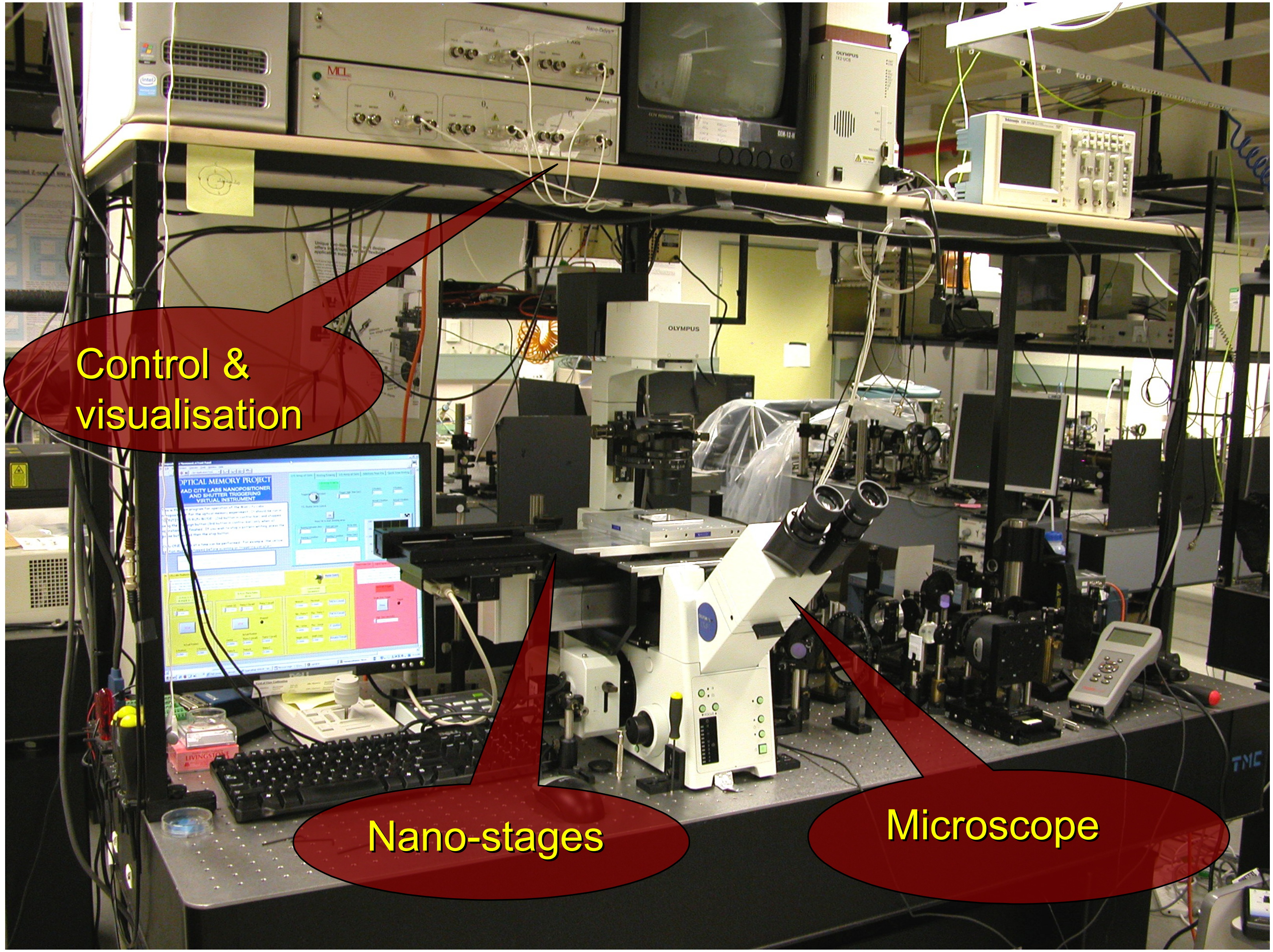


Warm Dense Matter
by confined micro-explosion

Spot size	**< 1 m**
Pulse duration	**~ 100 fs**
Pulse energy	**~ 200 nJ**

E.G. Gamaly , A. Vailionis, V. Mizeikis, W. Yang, A.V. Rode, S. Juodkazis,
High Energy Density Physics 8 (2012) 13-17
Warm dense matter at the bench-top: Fs-laser-induced confined micro-explosion

Experimental installation



Control & visualisation

Nano-stages

Microscope

Outline

Beam propagation in a medium with intensity modified optical properties

Ionization, pressure/temperature conditions in the energy absorbing volume

Shock wave formation and structure: light/heavy ions spatial separation

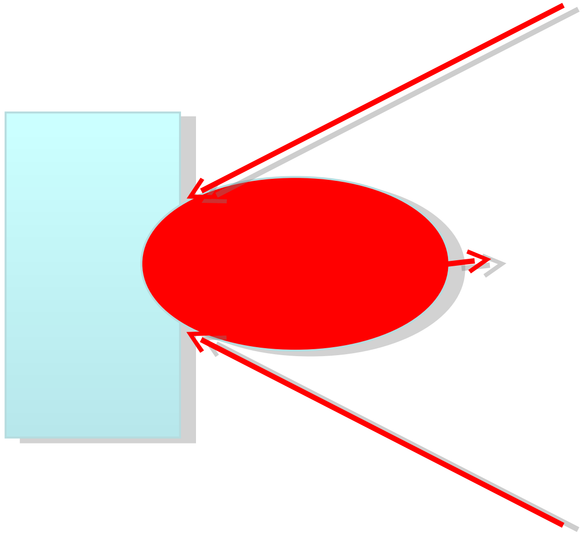
Rarefaction wave and void formation

Experiments: confined micro-explosion

at the boundary of transparent (silica) and opaque (Si) solids

Future directions

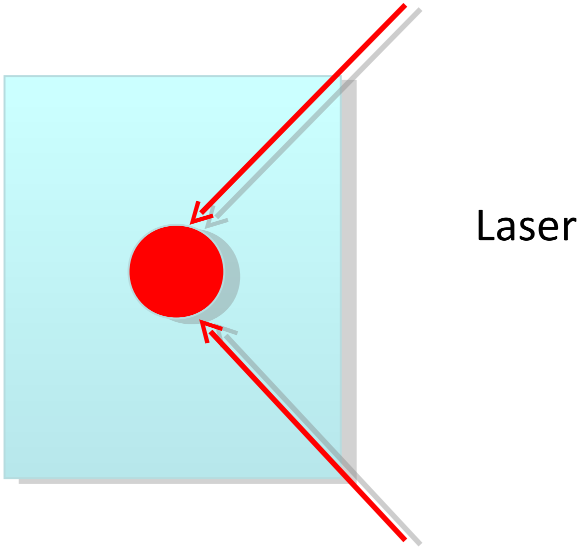
Laser – surface interaction



Absorbed energy shared between Internal energy and expansion

$$P_{max} \sim I^{2/3}$$

Confined interaction



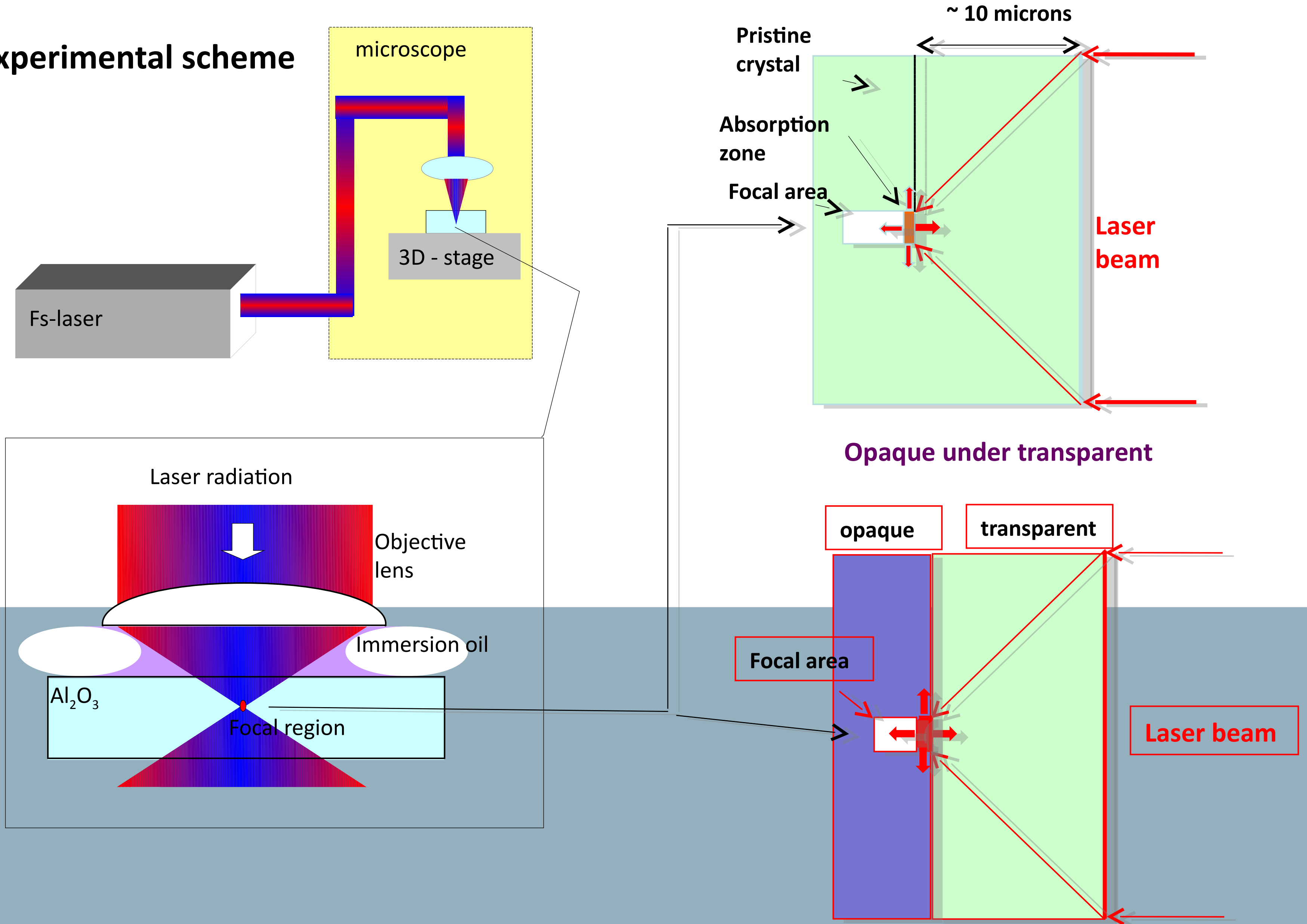
Whole absorbed energy is in the Internal energy

$$P_{max} \sim I \times t_{pulse}/l_{abs}$$

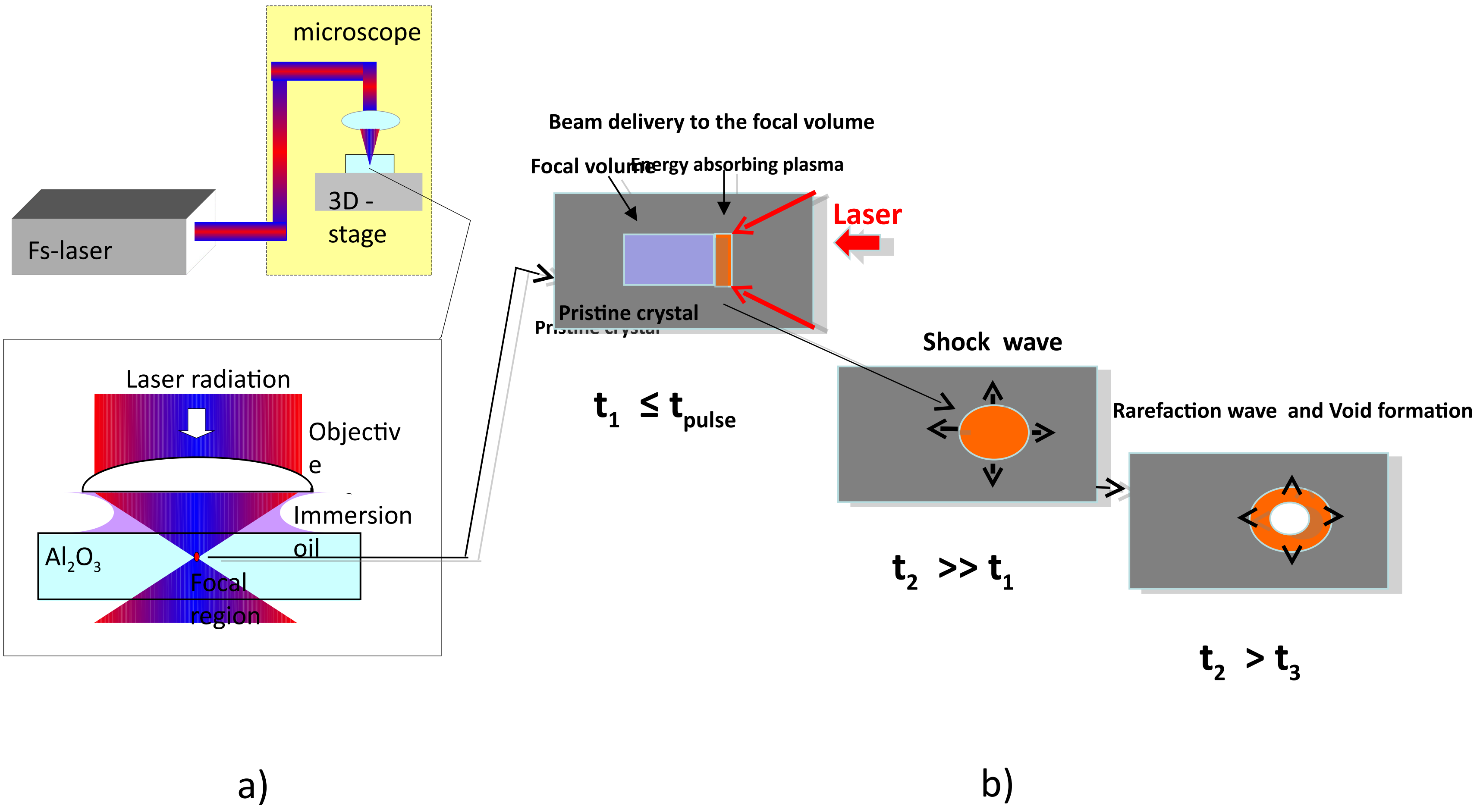
Energy carriers are massless

Micro-explosion in transparent solid and at opaque/transparent boundary

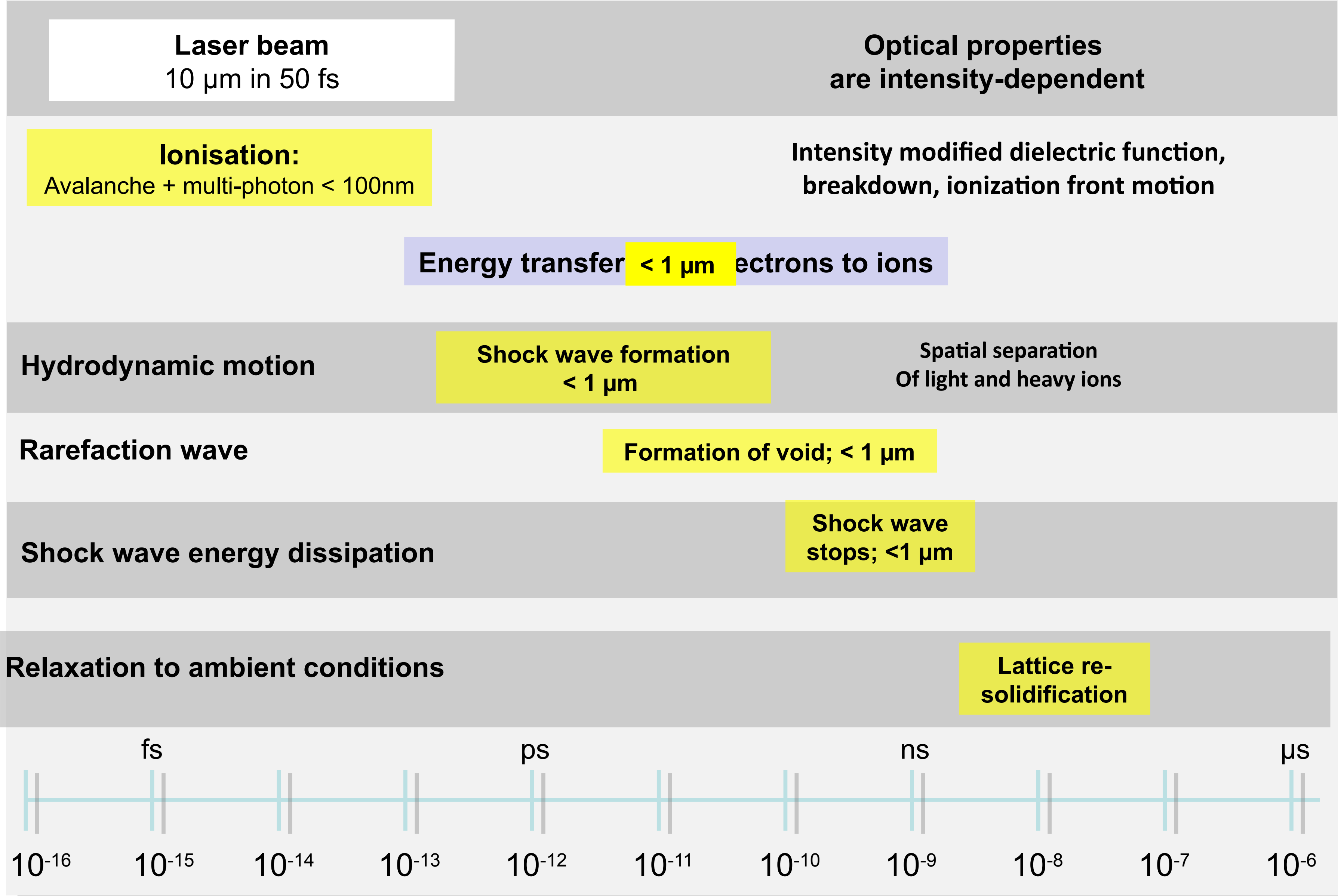
Experimental scheme



Time sequences for the stages of laser-induced micro-explosion



Micro-explosion: succession of processes, time and space scales



Laser beam
10 µm in 50 fs

Optical properties are intensity-dependent

Ionisation:
Avalanche + multi-photon < 100nm

Intensity modified dielectric function, breakdown, ionization front motion

Energy transfer **< 1 µm** **ectrons to ions**

Hydrodynamic motion

Shock wave formation < 1 µm

Spatial separation Of light and heavy ions

Rarefaction wave

Formation of void; < 1 µm

Shock wave energy dissipation

Shock wave stops; <1 µm

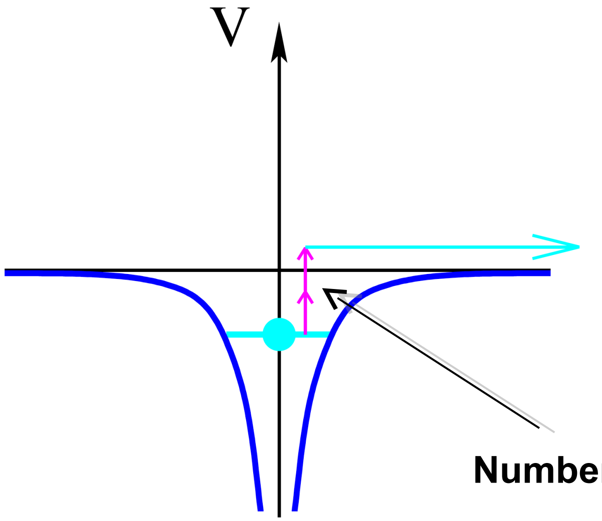
Relaxation to ambient conditions

Lattice re-solidification

fs ps ns µs

10^{-16} 10^{-15} 10^{-14} 10^{-13} 10^{-12} 10^{-11} 10^{-10} 10^{-9} 10^{-8} 10^{-7} 10^{-6}

Ionisation mechanisms

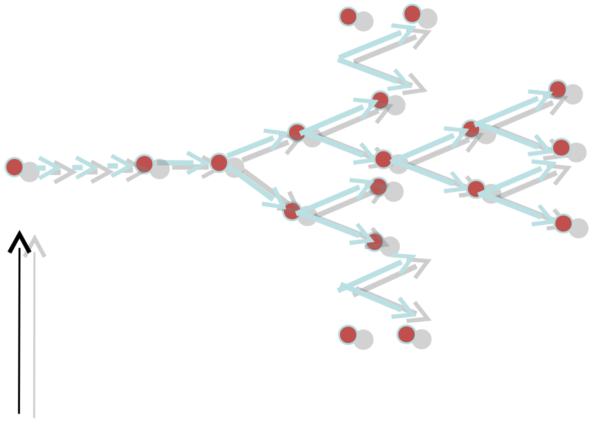


Number of phonons, n_{ph}

Multi-photon ionisation

$$w_{mpi} \quad \left(\frac{I}{\Delta_{gap}}\right)^{n_{ph}}$$

Ionisation by electron impact



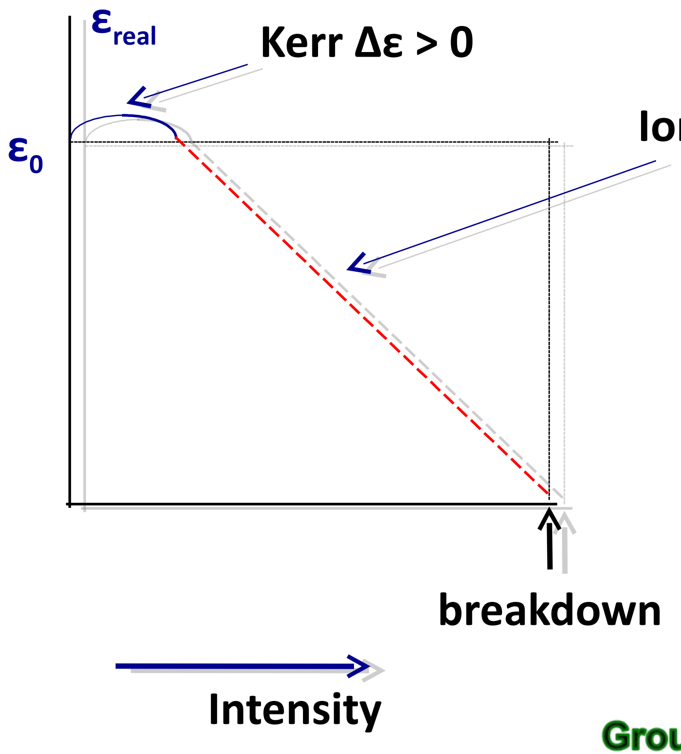
Avalanche ->Electron gains energy > band gap

$$w_{imp} \quad I/\Delta_{gap}$$

For silica and 800 nm (1.55eV) $n_{ph} \approx 5$

Intensity modified dielectric function: effects on beam propagation

$$P_{laser} < P_{cr}^{self.f} = 093l_0^2/(2p \quad n_0 n_2)$$



$$e \gg e_0 + De_{Kerr} - De_{ion}^{re} + iDe_{ion}^{im};$$

$$De_{ion}^{re} \qquad De_{ion}^{im} \qquad n_e/n_{cr}$$

Ionization effects on beam propagation

Defocusing

Group velocity increase and fall

$$\Delta v_g = \frac{c^2 w_{ope}^2}{w_0^2 v_{0g}} \frac{Dw}{w} - \frac{Dn_e}{n_{0e}}$$

Frequency blue shift

$$\Delta\omega \approx \frac{L}{2w_0 v_{0g}} \frac{dw_{pe}^2}{dt} = \frac{Lw_0}{2v_{0g} n_{cr}} \frac{dn_e}{dt}$$

Wave propagation in gradually ionized medium

$$\nabla \times \nabla \times E = -\frac{1}{c^2}\frac{\partial^2 D}{\partial t^2}\ ;\ D = \varepsilon E$$

$$divD = 0$$

3D Maxwell equations

$$\varepsilon = \varepsilon_{pol} + i\frac{4\pi\sigma}{\omega} = \varepsilon_{re}(n_e) + i\varepsilon_{im}(n_e)$$

intensity/temperature dependent dielectric function

$$\frac{\partial n_e}{\partial t} = W(I(r,z,t)) - R(r,z,t)$$

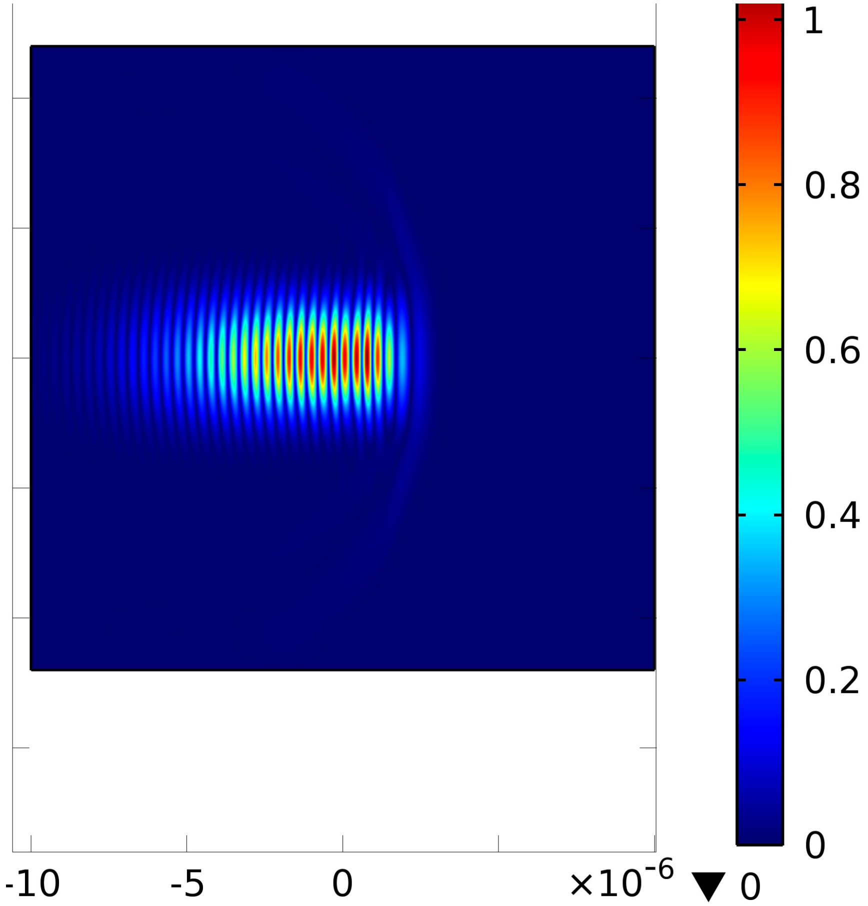
Rate equation for electrons: ionization minus recombination

$$\frac{\partial \mathrm{E}_e}{\partial t} = Q_{abs} - Q_{e-ph}$$

$$\frac{\partial \mathrm{E}_L}{\partial t} = Q_{e-ph}$$

Electron and lattice (ions) temperature equations

Electric field snapshot during the propagation (25 fs, Gauss, ionization)



Distance in microns

Condition for the optical breakdown threshold

Number density of electrons in conduction band

$$n_e\Big|_{breakdown} = n_c = \int_0^{t_{ion}} n_e(I(z,r,t))dt$$

Electronic rate equation

$$\frac{\partial n_e}{\partial t} = w_{imp} n_e + w_{mpi}(n_a - n_e) - R$$

Breakdown time, *t_{ion}*, is time for accumulation electron density equal to the critical one

10 fs ≤ t ≤ 100

Critical electron number density

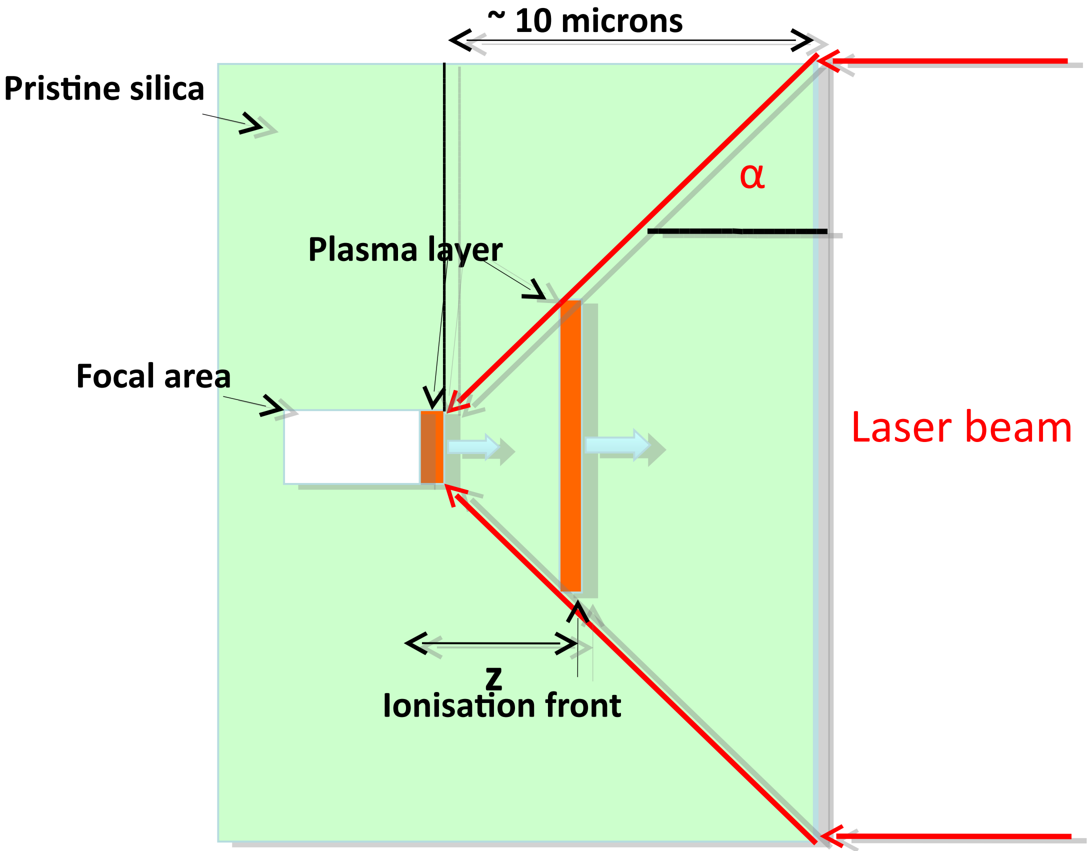
$$n_c = \frac{m_e \omega^2}{4\pi e^2}$$

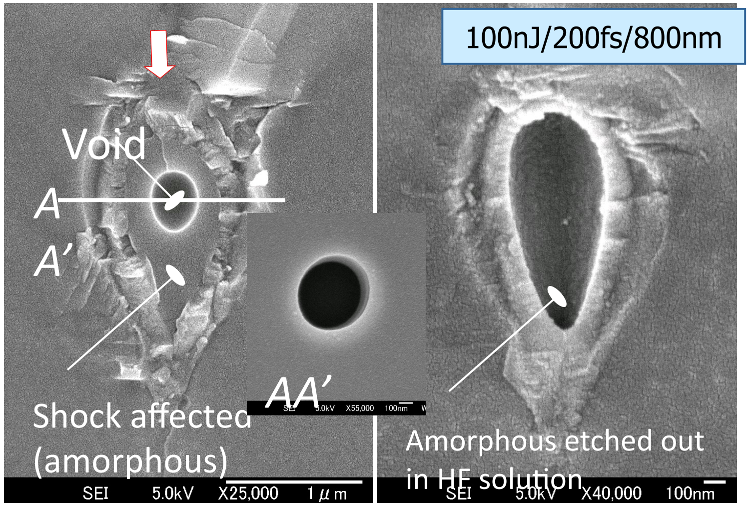
At breakdown spot $v_{group} = 0$; $\varepsilon_{real} \approx 0$; wave becomes evanescent

Ionization front (critical surface) motion

$$z(t_p) = \frac{r_f}{tga}\left(f^{1/2} - 1\right)$$

$$f = \frac{E_{las}(t_p)}{p r_f^2 F_{thr}}$$





S. Juodkazis, ***et.al.***, Phys. Rev. Lett. 96, 166101 (2006)

E. G. Gamaly, *et al., Phys. Rev. B*, 73, 214101 (2006).
2. S. Juodkazis, *et al.,* Appl. Phys. Lett. 88, 201909 (2006).

Australian
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University

Maximum pressure and electron temperature in the absorption volume

Total deposited energy
(hot electrons, cold ions)

$$E_{dep} = \frac{2AF_p}{l_{abs}}; \quad F_p(r,z,t) = \overset{t_p}{\underset{0}{}} I_0(r,z,t)dt$$

Maximum pressure driving a shock wave in sapphire

$$P_{\max} = \frac{E_{dep}}{V_{abs}} » (3 \quad 4)TPa >> strength \;\; of \;\; any \;\; material$$

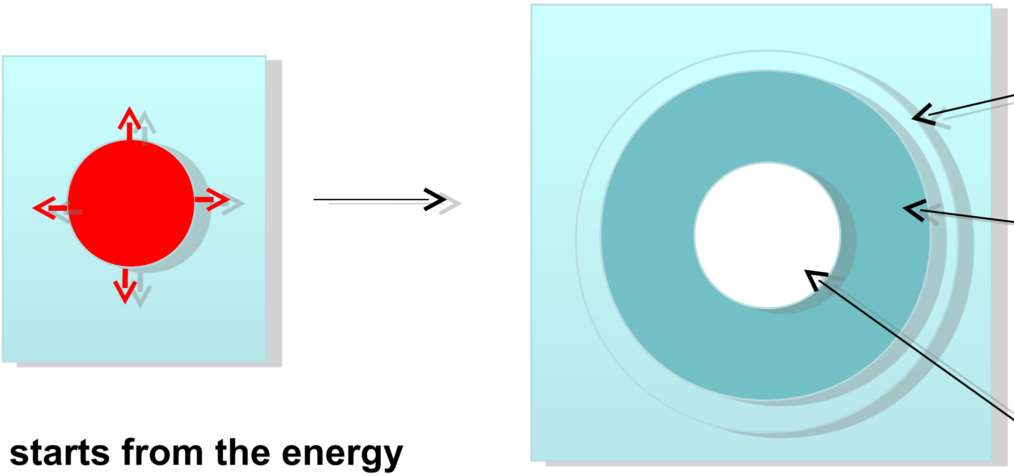
$$\boldsymbol{V_{abs}} » p\boldsymbol{r}^2_{foc}\boldsymbol{l}_{abs}$$

(E_{Las} = 10^{-7} J, F_p = 70.4 J/cm^2; $S_{1/2}$= 0.142 m^2, l_{abs}= 65 nm;

V_{abs}= 10^{-2} m^3 ; A = 0.62)

A. Vailionis, E. G. Gamaly, V. Mizeikis, Wenge Yang, A. V. Rode & S. Juodkazis, Nature communications (2011) | DOI: 10.1038/ncomms1449
"Evidence of super-dense aluminium synthesized by ultrafast micro-explosion

Absorbed energy density and pressure from conservation law



Shock converted to sound wave, r_{stop}

Material pulled off from the void

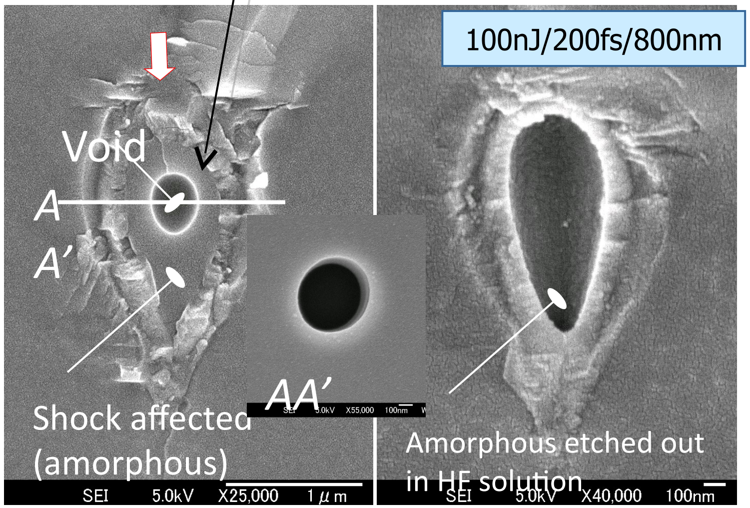
void

SW starts from the energy absorption zone

Energy conservation

$$\frac{4}{3}\pi Y_{sapp}\, r_{stop}^3 \approx E_{abs}$$

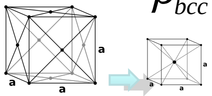
Experiment in sapphire

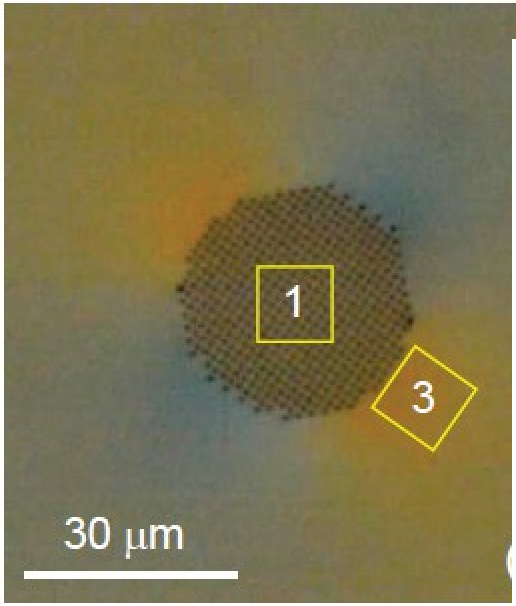


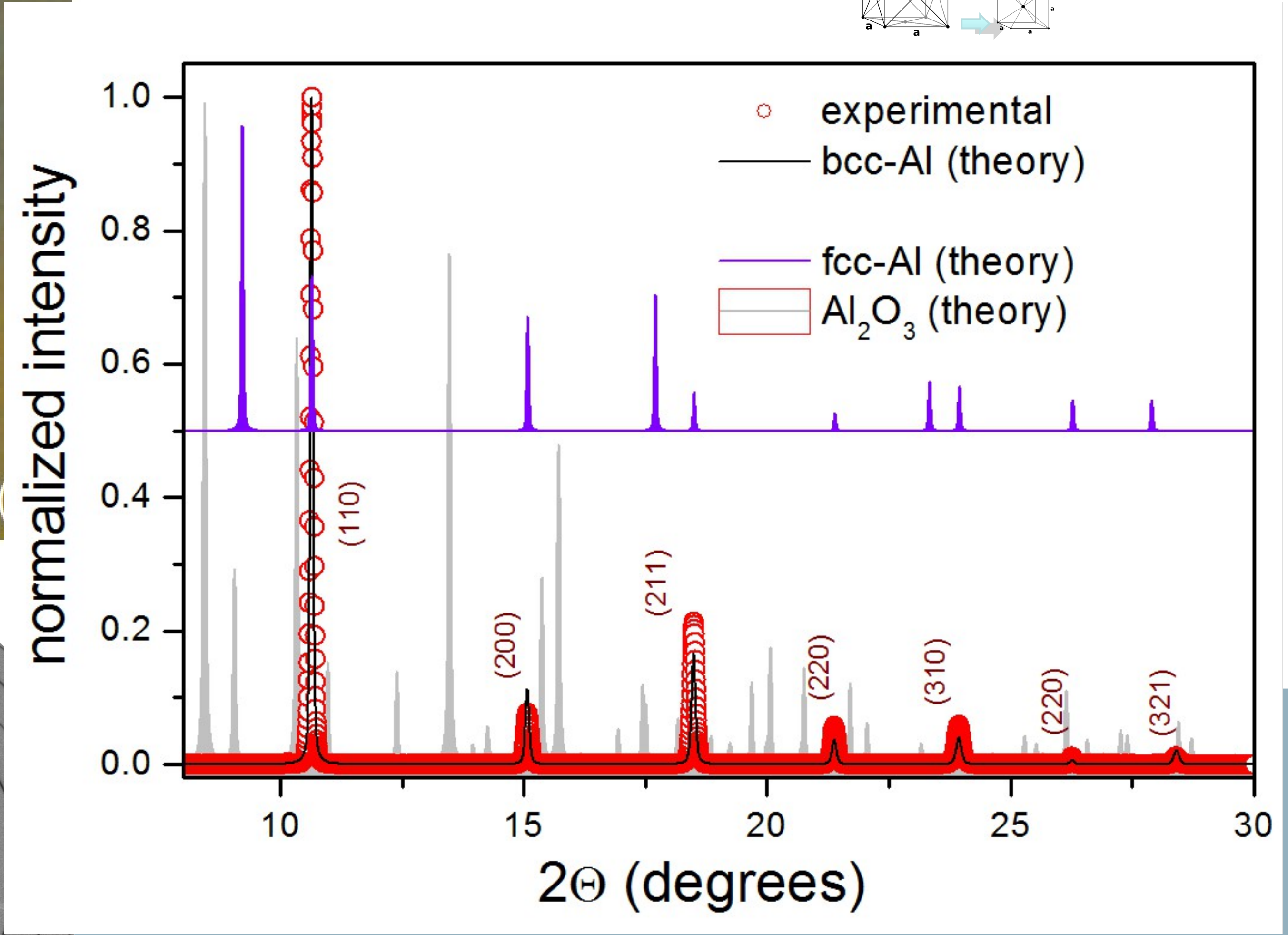
Discovery of bcc-Al by Synchrotron X-ray diffraction

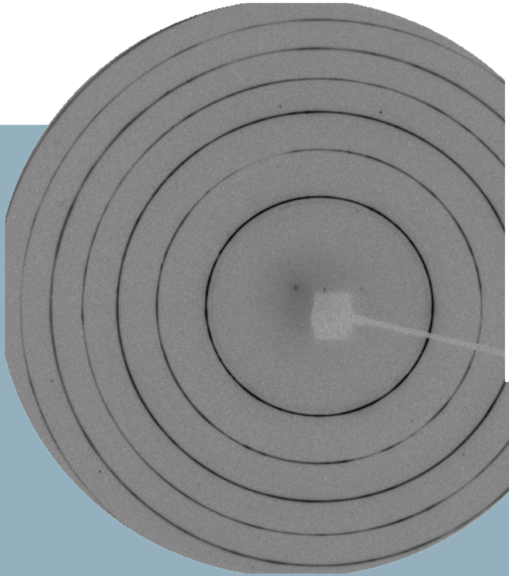
(collaboration with Argonne APS, 2-ID-D)

ρ_{bcc} » $3.78\,g/cm^3$









A. Vailionis, E. G. Gamaly, V. Mizeikis, Wenge Yang, A. V. Rode & S. Juodkazis,
B. Nature communications (2011) | DOI: 10.1038/ncomms1449
“Evidence of super-dense aluminium synthesized by ultrafast micro-explosion

Discovery of bcc Al inside sapphire

fcc-Al hcp-Al (120-360 GPa) **bcc-Al (200-560 GPa)**

$$a_{bcc} = a_{fcc}/\sqrt{2} = 2.865\ \text{Å}$$

Size of bcc-Al crystallite: 18 ± 2 nm

Spatial separation of Aluminium and Oxygen (?)

Preserved stochiometry of Al_2O_3 ;

all laser-affected material confined inside a crystal

A. Vailionis, E. G. Gamaly, V. Mizeikis, Wenge Yang, A. V. Rode & S. Juodkazis, Nature communications (2011) | DOI: 10.1038/ncomms1449
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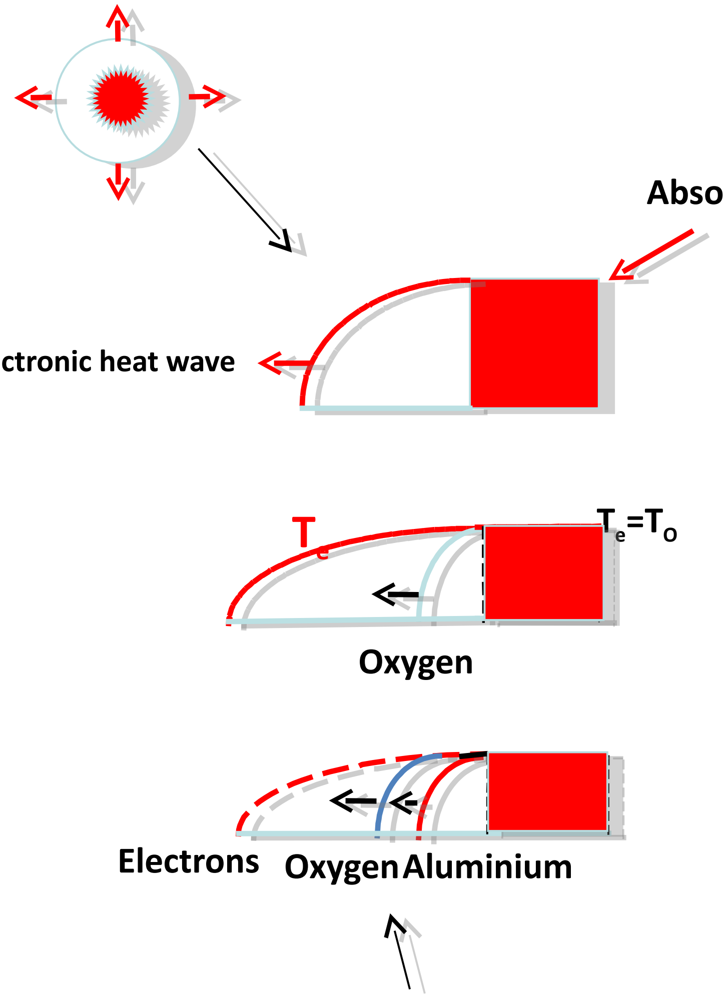
Formation and structure of the shock wave front

Electrons, oxygen and aluminium ions start moving in different time with different velocities $v_e >> v_O > v_{Al}$

$$t^{energy}_{e-ion} = M_{ion} / m_e n_{e-i}$$



$t < t^{energy}_{e-Al,O}$

$t \quad t^{energy}_{e-O}$

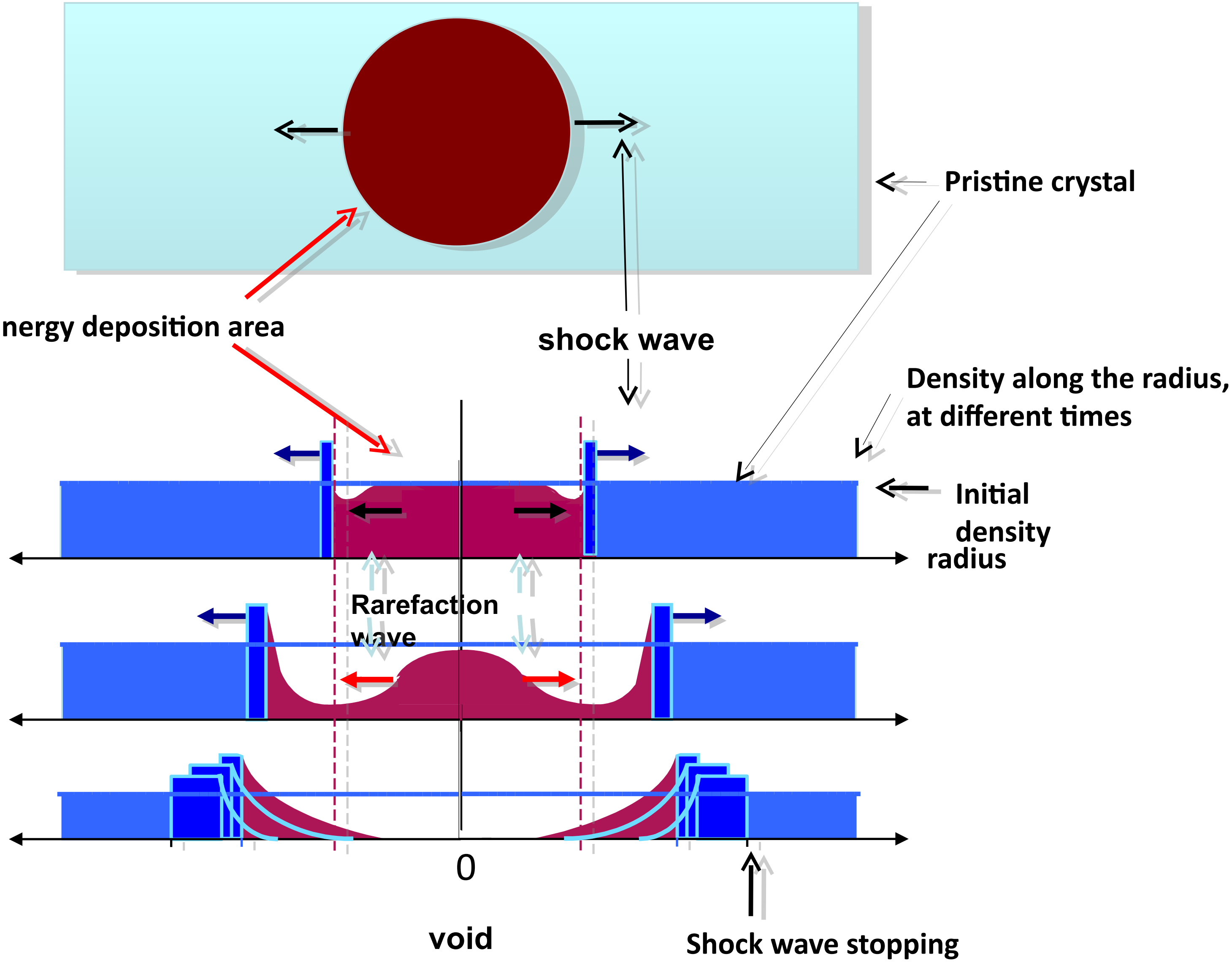
Electrons transferred energy to oxygen

$t \quad t^{energy}_{e-Al}$

Electrons transferred energy to Aluminium

$\Delta x_{separation} \qquad M_l^{1/2} T^2 (M_h / M_l - 1)$

Energy deposition, shock and rarefaction wave formation and stopping



Energy deposition area

shock wave

Pristine crystal

Density along the radius, at different times

Initial density

radius

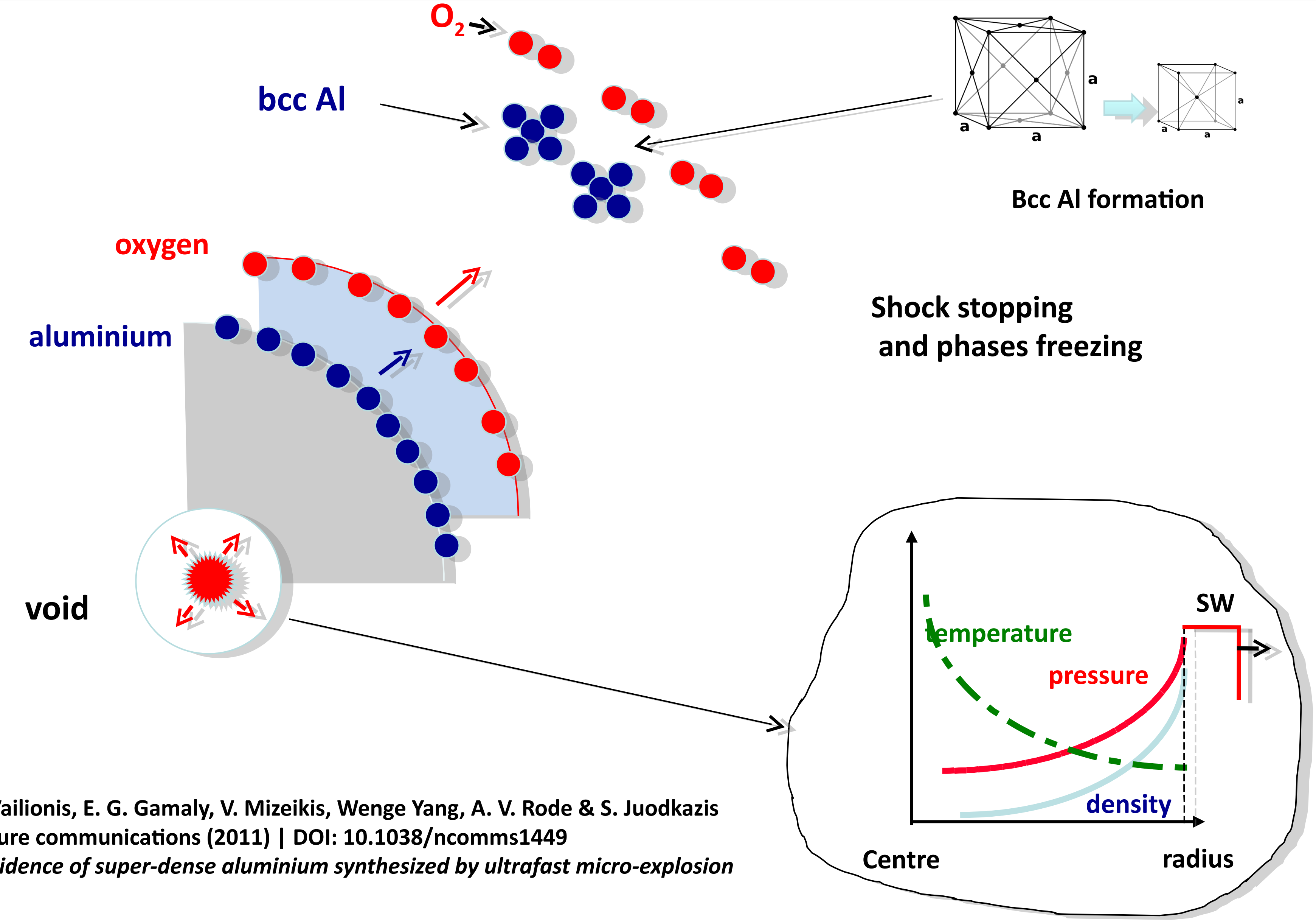
Rarefaction wave

0

void

Shock wave stopping

Shock wave and void generation, light/heavy ions separation



Bcc Al formation

Shock stopping
and phases freezing

A. Vailionis, E. G. Gamaly, V. Mizeikis, Wenge Yang, A. V. Rode & S. Juodkazis
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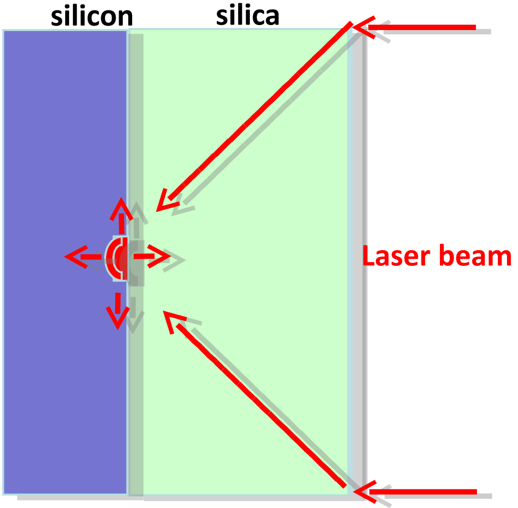
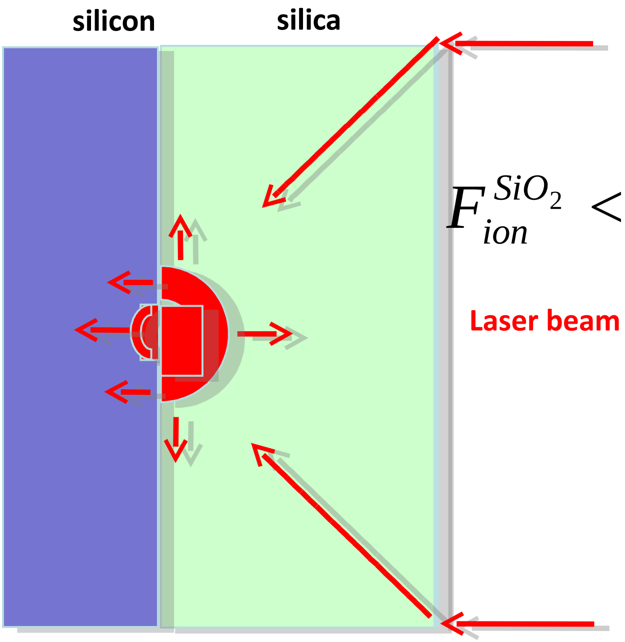
Shock waves formation at the transparent/opaque boundary



$$F_{ion}^{SiO_2} = F_{las}$$

$$\frac{L_{shock}^{transp}}{L_{shock}^{opaque}} \gg {\frac{Y_{opaque}}{Y_{transp}}}^{1/3}$$

$$Y_{si} \approx 2.2\, Y_{SiO2}$$

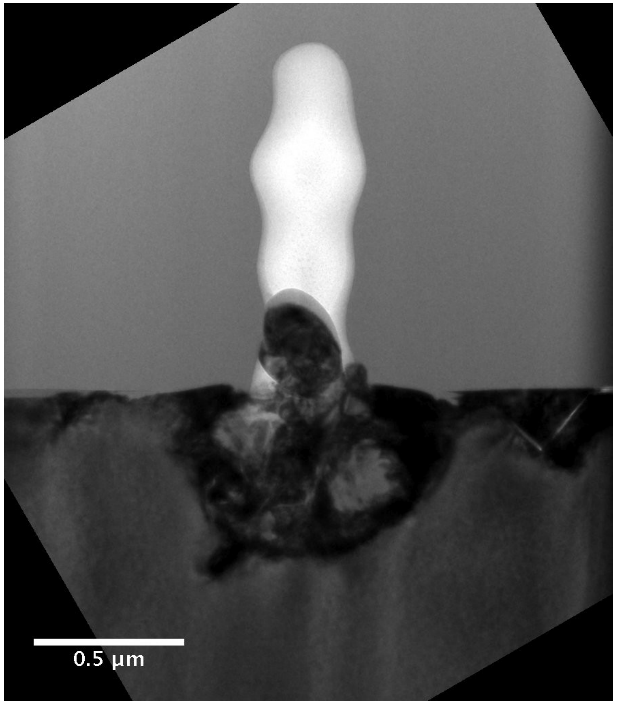


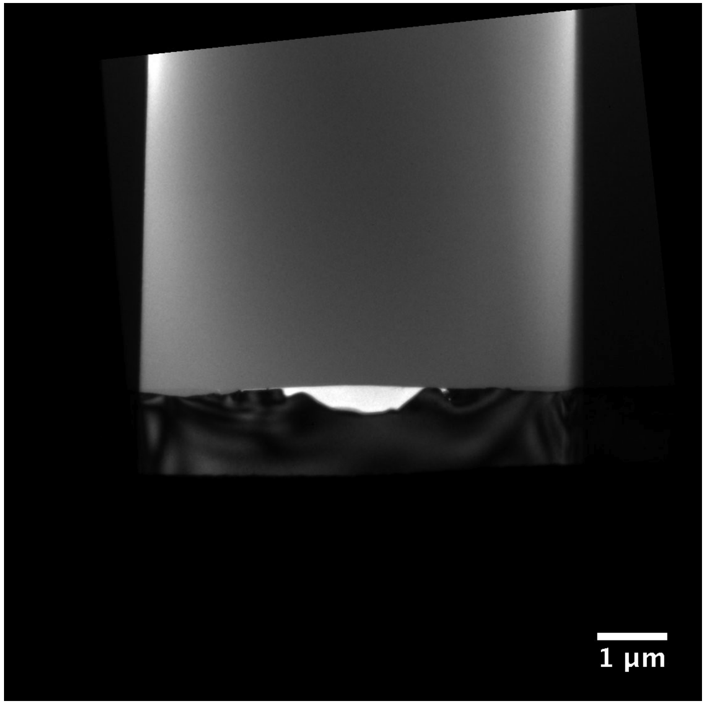
$$F_{ion}^{SiO_2} < F_{las}$$

Conditions for the maximum energy deposition in the opaque medium:

$F_{las} \leq F_{ion\ transp}$; $Y_{transp} \approx Y_{opaque}$

Micro-explosion at Silica/Si interface at different laser fluences





radius = 0.368 µm
Fluence = 95 J/cm2

radius = 3.127 µm

Fluence = 2.6 J/cm2

Future studies

Olivine – separation of iron

Diamond – search for C8

High pressure phases of Silicon

High pressure phases of metals in metal/oxide combinations

Transparent oxides of heavy metals

Femtosecond pump-probe

micro-explosion in transparent crystals: stishovite – high pressure phase of silica, BaF_2, CaF_2…

Olivine $(MgFe)_2SiO_4$ - one of the most common minerals on Earth, Moon and Mars

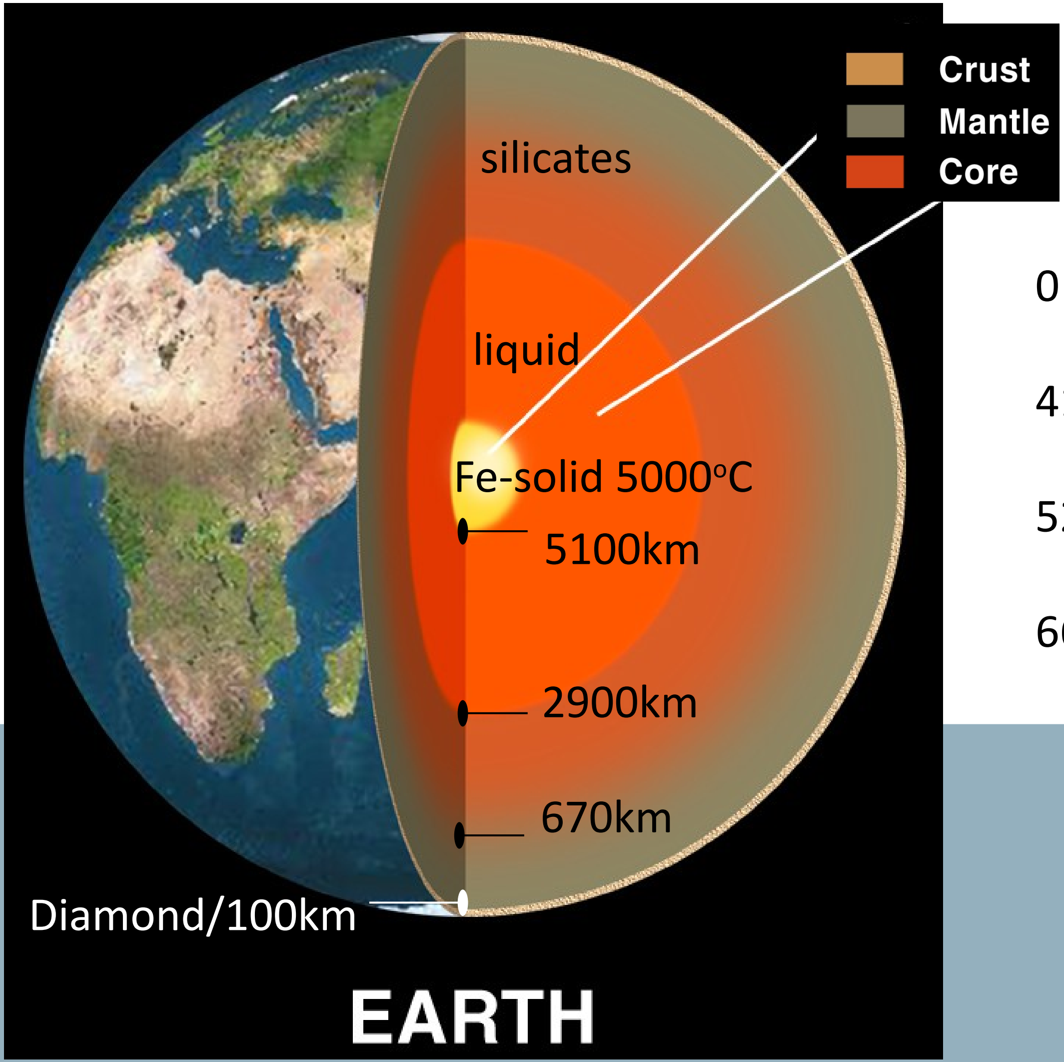
Separation of iron from the other elements in olivine
By micro-explosion

Iron + Nickel core of Earth exists at pressure 330 Gpa

= sum of thermal and gravitational pressure

Speculation: Modelling Formation of Earth iron core from Olivine?

Pressure temperature (p,T) in the inner Earth



Reflection of seismic waves occurs at 410, 520, and 660 km.

0 km/ 10^5Pa olivine (Mg,Fe-silicate)

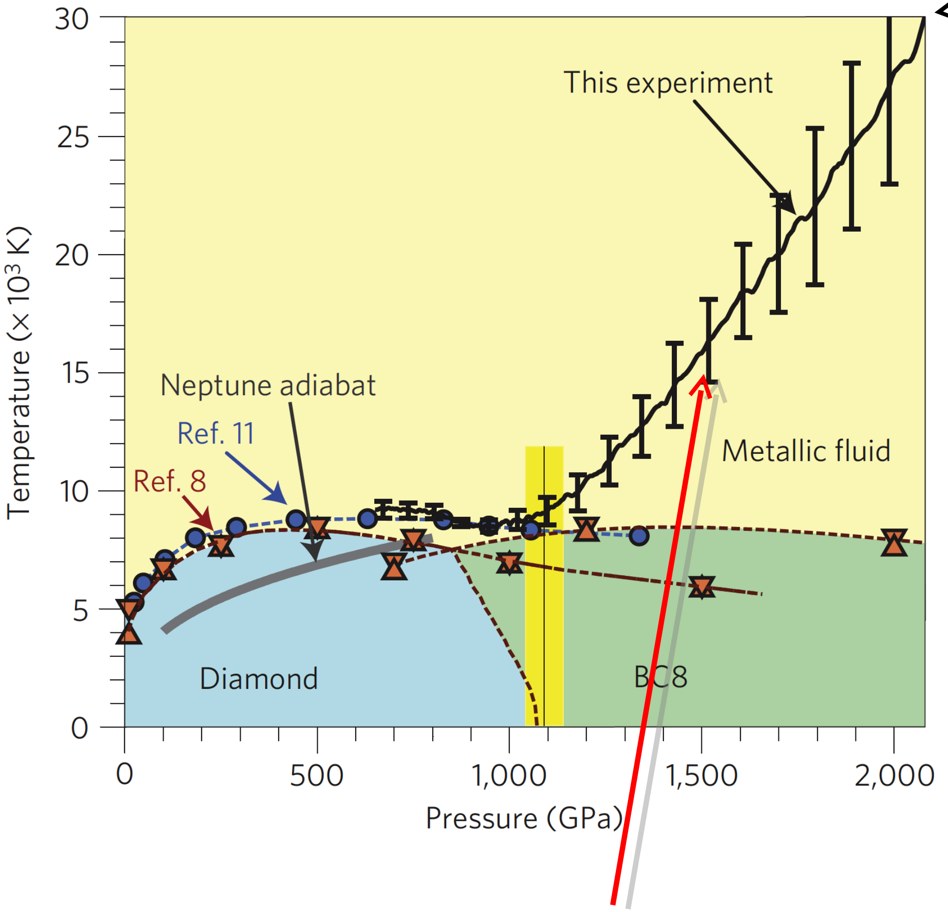
410 km/ 14 GPa wadsleyite

520 km/ 18 GPa ringowoodite

660 km/ 23 GPa perovskite or magnesiowüstite

1.3×10³ J, 1 nanosecond laser - surface interaction

shock wave compression and expansion



BC8, in micro-explosion?

8×10^{-7} J, 100 fs, confined micro-explosion

Melting temperature of diamond at ultrahigh pressure

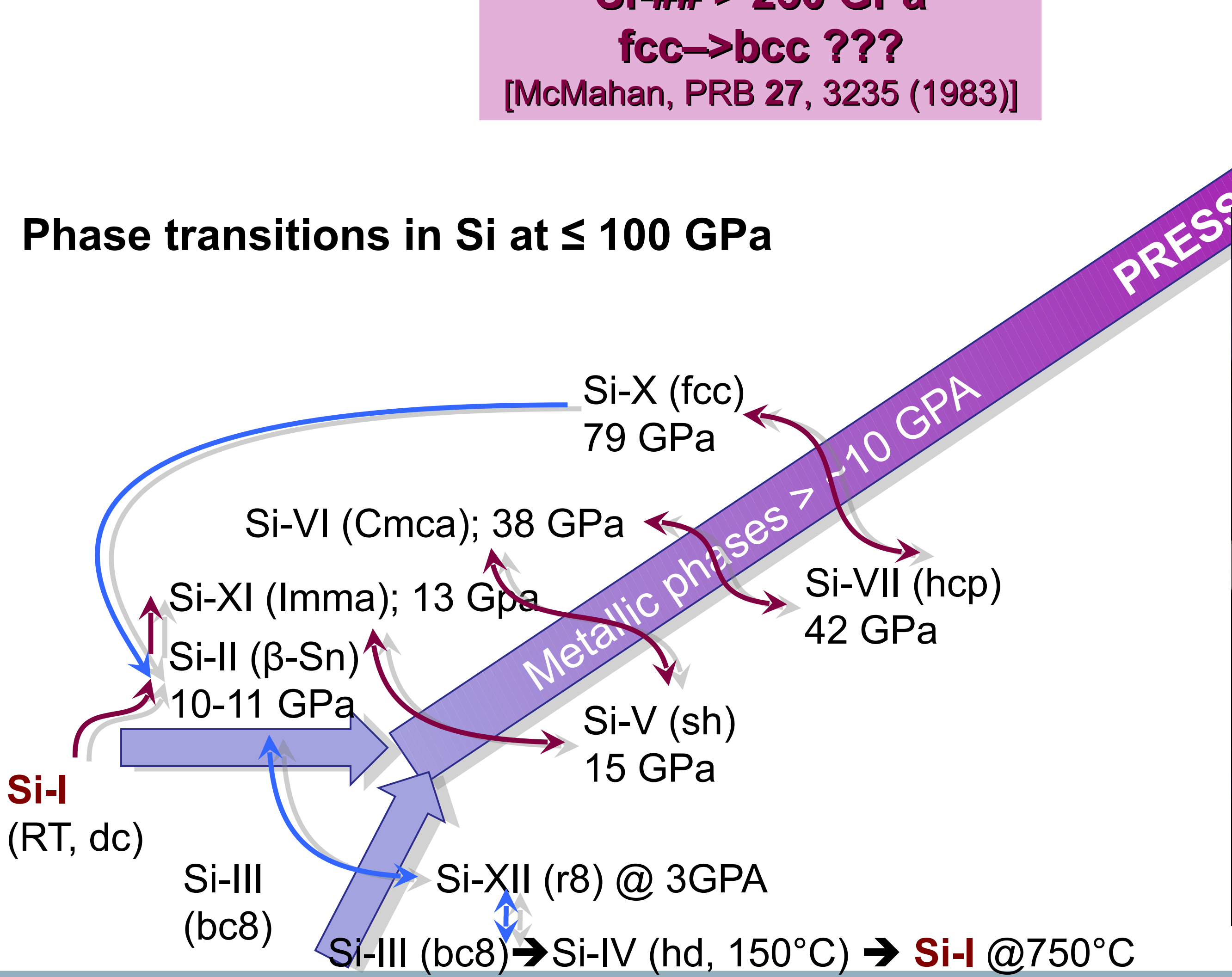
J. H. Eggert, D. G. Hicks, P. M. Celliers, D. K. Bradley, R. S. McWilliams, R. Jeanloz, J. E. Miller, T. R. Boehly and G.W. Collins
NATURE PHYSICS VOL 6 , JANUARY 2010

High pressure phases of Silicon

Search for formation of new high pressure phases of Silicon By confined micro-explosion

Si-## > 250 GPa
fcc–>bcc ???
[McMahan, PRB 27, 3235 (1983)]

Phase transitions in Si at ≤ 100 GPa



PRESSURE

Metallic phases > ~10 GPA

Si-X (fcc)
79 GPa

Si-VI (Cmca); 38 GPa

Si-VII (hcp)
42 GPa

Si-XI (Imma); 13 Gpa

Si-II (β-Sn)
10-11 GPa

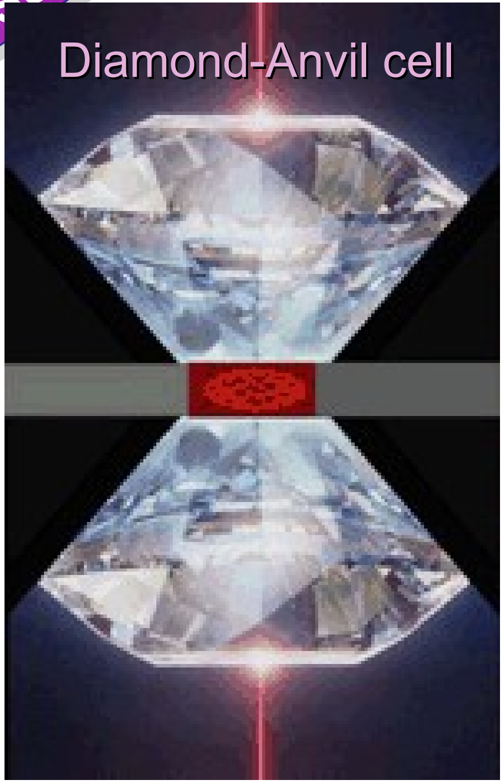
Si-V (sh)
15 GPa

Si-I
(RT, dc)

Si-III
(bc8)

Si-XII (r8) @ 3GPA

Si-III (bc8)➔Si-IV (hd, 150°C) ➔ **Si-I** @750°C



Diamond-Anvil cell

Conclusions

Generation of pressure in excess 200 GPa by micro-explosion in opaque Silicon buried under transparent silica layer

Observation of amorphous Si of unknown structure evidenced by the unconventional Raman peaks

Establishing optimum conditions for maximizing the energy deposition in opaque medium

Effects of ion front motion in direction opposite to the laser beam on the energy density

Increase in the Coulomb interactions enhances light/heavy ion separation effect

Future studies: new materials; *in situ* diagnostics

Collaboration team

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The Australian National University

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Electronic Materials Engineering, Research School of Physics and Engineering,
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Wenge Yang, *Argonne National Laboratory, USA*

Vito Roppo, *CNRS, Paris, France*

Thank you !

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High Energy Density Physics 8 (2012) 13-17
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2. S. Juodkazis, *et al.,* Appl. Phys. Lett. 88, 201909 (2006).
3. S. Juodkazis, *et.al.*, Phys. Rev. Lett. 96, 166101 (2006).

Lena Bressel, Dominique de Ligny, Eugene G. Gamaly, Andrei V. Rode, Saulius Juodkazis,
Observation of O_2 inside voids formed in GeO_2 glass by tightly-focused fs-laser pulses,
Optical Material Express, September 2011

-Quartz and silica converts to stishovite (4.29 g/cm^3)
- in the range between ~30 Gpa-110 Gpa.

Silica and stishovite melts at P > 110 GPa >> shear modulus for liquid silica ~ 10 GPa

-New phases formed inside the bulk SiO_2 (probably -stishovite 4.29 g/cm^3 in the range between ~30 -110 Gpa, 5-7% of the shell mass)
-Dense phase: Nano-crystallites, nano-clusters?

-The heating rate by powerful short pulse laser ~ 50 eV/200 fs = 3x10^{17}Kelvin/s

--The cooling rate ~ 50 eV/2ns ~ 3x10^{14}K/s