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# Ultrafast dynamics in matter under extreme conditions

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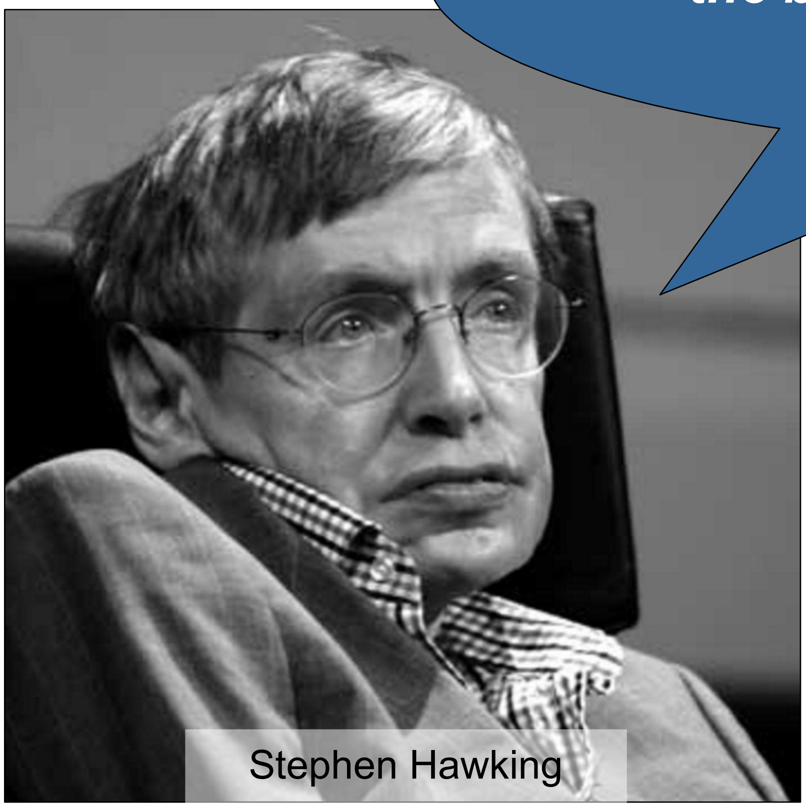
# Extreme conditions

*"We already know the laws that govern the behavior of matter under all but the most extreme conditions" **

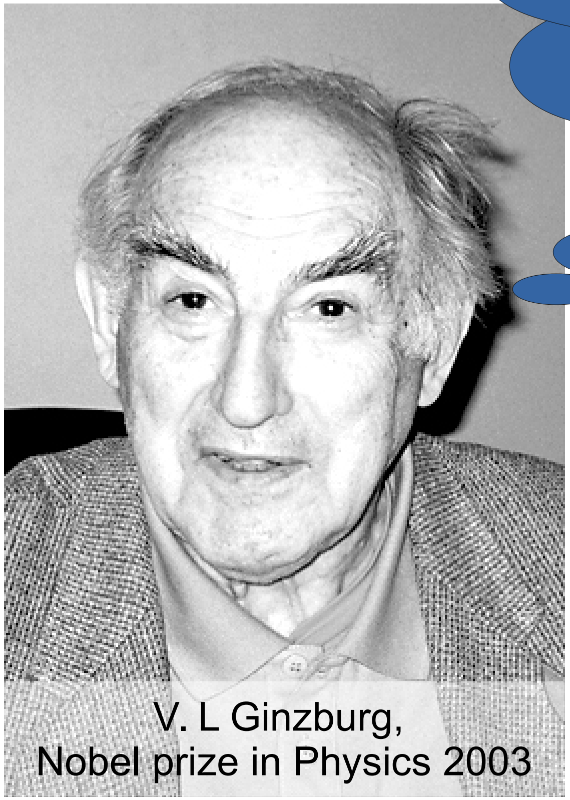


Stephen Hawking

*) S. Hawking. A Brief History of Time (p. 168).

# High-energy-density (HED) physics

Half of the 30 problems of “the physics minimum at the beginning of the XXIst century” are to a greater or lesser degree dedicated to high-energy-density physics.*



V. L Ginzburg,
Nobel prize in Physics 2003

*) V. L. Ginzburg, Phys. Usp. 47(11), 1155 (2004)

# Extreme conditions and HED physics

- High energy densities ⟷ high temperatures and densities ⟷ extreme conditions
- Recent outstanding experiments revealing the Higgs Boson and the gravitational waves were associated with high-energy-density events
- Extreme conditions:
  - energy densities exceeding $10^4$ –$10^5$ J/cm$^3$ (typical binding energy of condensed matter)
  - pressure level of Mbar
- The major part (90–95 %) of baryon (visible) matter in Universe is under extreme conditions

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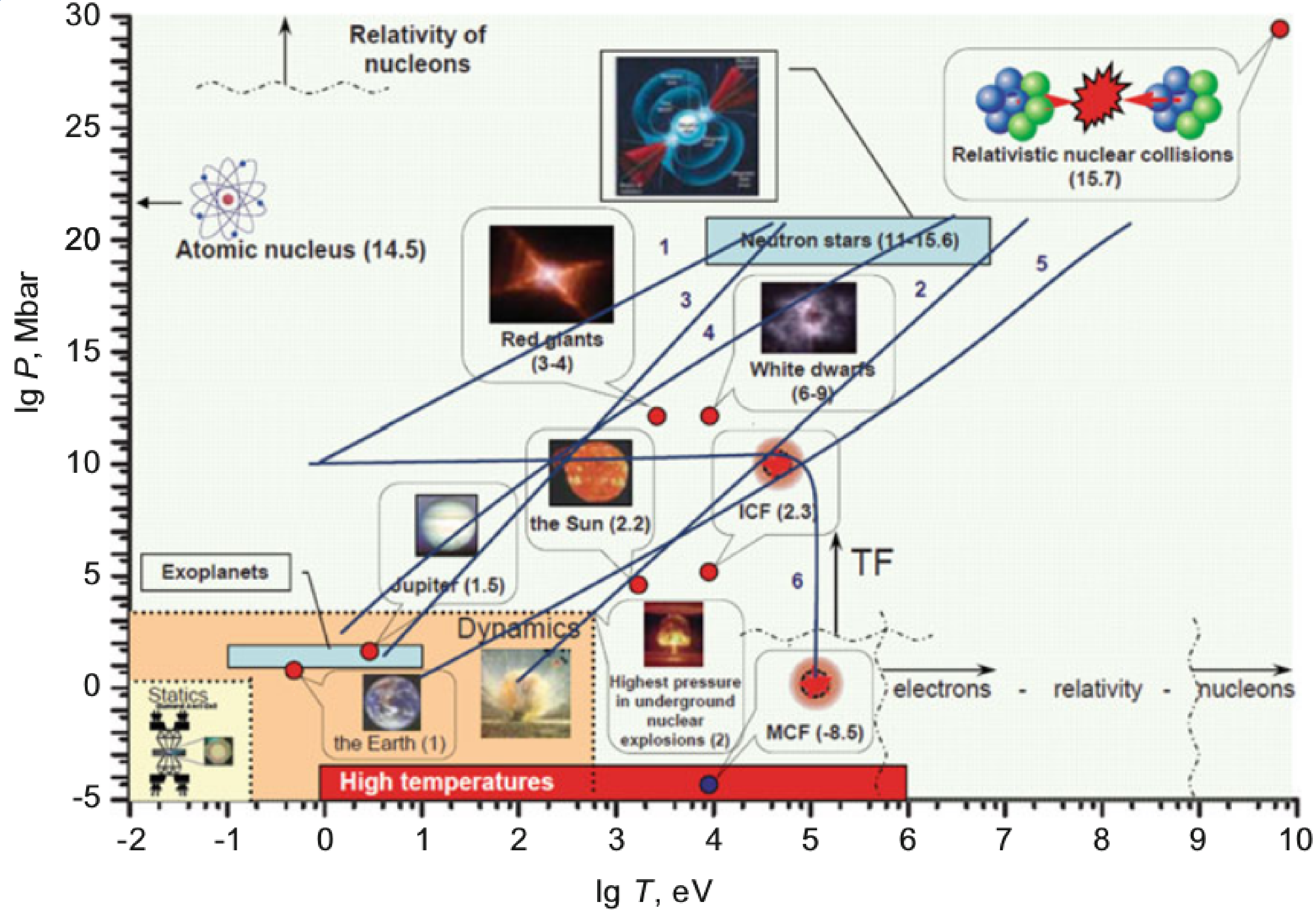
# Extreme conditions in the universe

| | Pressure | Temperature | Density |
|---|---|---|---|
| Mariana Trough | 1.2 kbar | ... | ... |
| Earth's center | 3.6 Mbar | 0.5 eV | 10–20 $g/cm^3$ |
| Jupiter's center | 40-60 Mbar | 2 eV | 30 $g/cm^3$ |
| Sun's center | 240 Gbar | $1.6 \cdot 10^3$ eV | 150 $g/cm^3$ |
| Inertial plasma confinement | 200 Gbar | $10^8$ eV | 150-200 $g/cm^3$ |
| White dwarfs | $10^4$-$10^9$ Tbar | $1.6 \cdot 10^3$ eV | $10^6$–$10^9$ $g/cm^3$ |
| Neutron stars | $10^{13}$-$10^9$ Tbar | $10^4$ eV | $10^{11}$ $g/cm^3$ |
| LHC | $10^{18}$ Tbar | $10^{10}$ eV | $10^{16}$ $g/cm^3$ |

1 eV = 11605 K

# Extreme states




From V. E. Fortov, “Extreme States of Matter”, Springer 2015

# How to access extreme conditions?

- How to access unexplored regions of the p-T phase diagram in the laboratory?

**a)** Compress the sample volume to increase the energy density ( $p=E/V$ )

**b)** Deposit a huge amount energy into the sample ( $E = pV$ )

- Static conditions (mechanical pressure):
  - Diamond anvil cells, DAC (5 Mbar)

- Dynamic conditions $10^{-6}$-$10^{-10}$ s (pulsed cumulation of high energy densities in substances):
  - lasers,
  - intense shock waves,
  - charged-particle beams,
  - high-current Z-pinches,
  - explosion devices,
  - multistage light-gas guns

# Laser driven extreme conditions

- In the laboratory, one desires that
  - the total amount of energy deposited into the sample is *completely* and *homogeneously* adsorbed in an excited volume;
  - the excited volume is small, in order to maximize the energy density;
  - the deposited energy remains in the excited volume for a time compatible with the dynamics under investigation;
  - the excited volume can be probed after a time delay after the onset of the extreme conditions.

- Such conditions can be fulfilled through laser heating
- In particular, if one studies lattice and electron dynamics in the sub-ps time scale for condensed matter, HED fs pulsed lasers are an extremely valid choice for generating extreme conditions

- An important advantage of sub-ps laser heating is the resulting **isochoric heating:**
  - sample density is constant
  - negligible energy transport
  - homogeneous thermodynamic conditions (when definable)
  - samples are not contaminated (high vacuum conditions, self-standing)

# Critical electron density

- When lasers are used to generate extreme conditions the wavelength ($\lambda$) plays a crucial role
- In condensed matter and dense plasma typically $\boldsymbol{n_e}$ > **$10^{22}$-$10^{23}$ cm$^{-3}$**

- The '*critical electron density*': $\boldsymbol{n_{cr}}$ **= $10^{29}$ cm$^{-3}$ / $\boldsymbol{\lambda^2}$ (Å)**
  - light can not propagate if $n_e > n_{cr}$ (plasma opacity)

- If $\boldsymbol{\lambda}$ **≥ 250 nm** (visible) → $\boldsymbol{n_{cr}}$ **≤ 1.6 * $10^{22}$ cm$^{-3}$**
  - optical lasers can not propagate in dense plasma ($n > 10^{22}$ cm$^{-3}$)
  - bulk heating is provided by ballistic electrons: poor temperature homogeneity

- If $\boldsymbol{\lambda}$ **≤ 50 nm** (EUV) → $\boldsymbol{n_{cr}}$ **≥ 4 * $10^{25}$ cm$^{-3}$**
  - EUV lasers can propagate!
  - Isochoric uniform heating is accessible by FELs

# Optical field ionization

- Optical field ionization plays an important role for the plasma formation when intense laser pulses interact with matter. To characterize the ionization processes, it is useful to introduce the Keldysh parameter $\gamma$ :

$$\gamma = \sqrt{\frac{I_P}{2U_P}}$$

Ionization potential

ponderomotive potential $U_P \approx I\lambda^2$

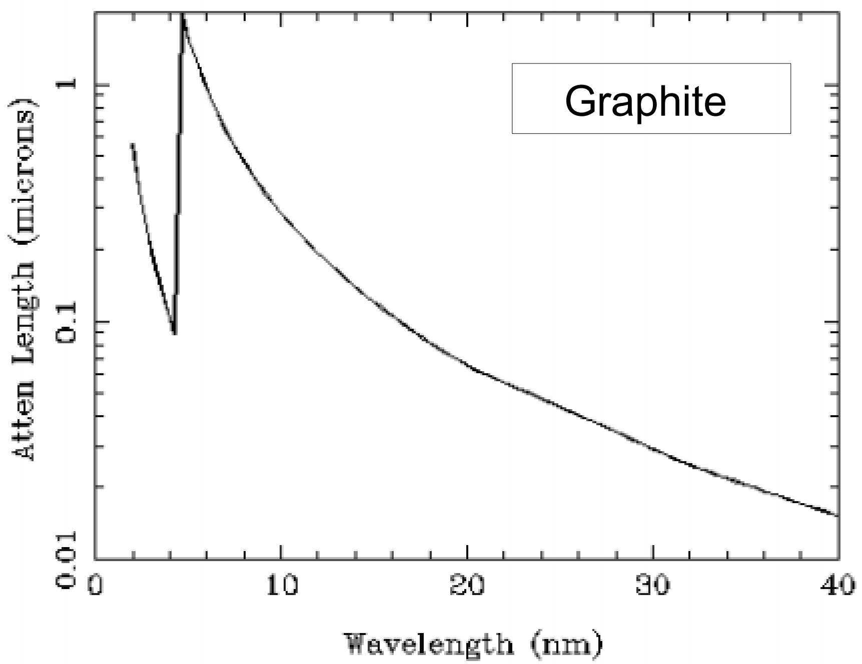
A ponderomotive force is a nonlinear force that a charged particle experiences in an inhomogeneous oscillating electromagnetic field.

- For $\gamma > 1$, the electric field of the laser can be considered as a small perturbation of the Coulomb field seen by the bound electrons (weak radiation field). Single photon ionization is dominant.
- For $\gamma << 1$ the field of the laser significantly affects the Coulomb potential of the atom and a perturbative treatment is not possible, anymore (strong radiation field). Electrons can escape from the atom by tunnel ionization. Multiphoton ionization is dominant.
- Under the same intensity ( $I$ ) conditions the ponderomotive potential is smaller for EUV than VIS light, therefore in order to avoid undesired effects such as huge multiphoton absorption, EUV is recommended.
- Moreover, EUV radiation can be absorbed by core electrons

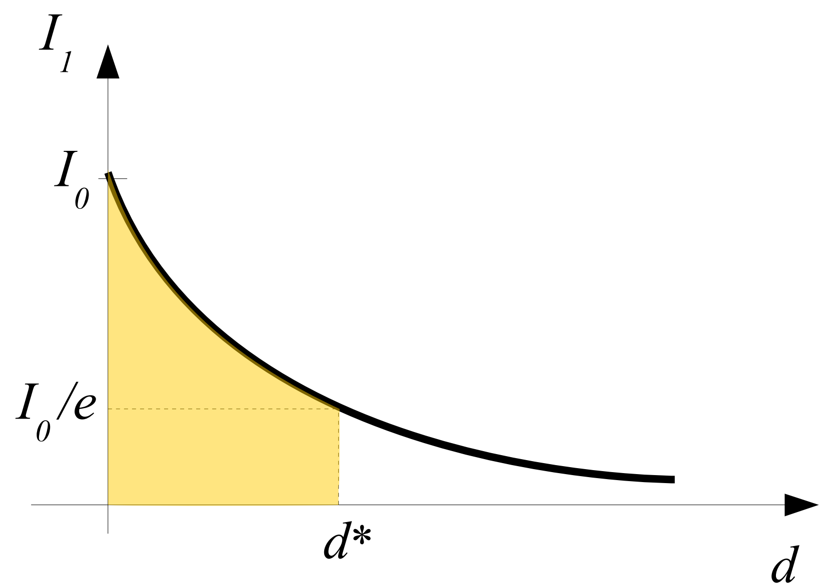
# Attenuation length

Under weak radiation field regime the amount of deposited optical energy into the sample can be estimated by using the Lambert-Beer equation

$$I_1(d,E) = I_0(E)e^{-\mu(E)d}$$




Attenuation Length ($d^*$):
The depth into the material measured along the surface normal where the intensity of x-rays falls to 1/e (~0.37) of its value at the surface.



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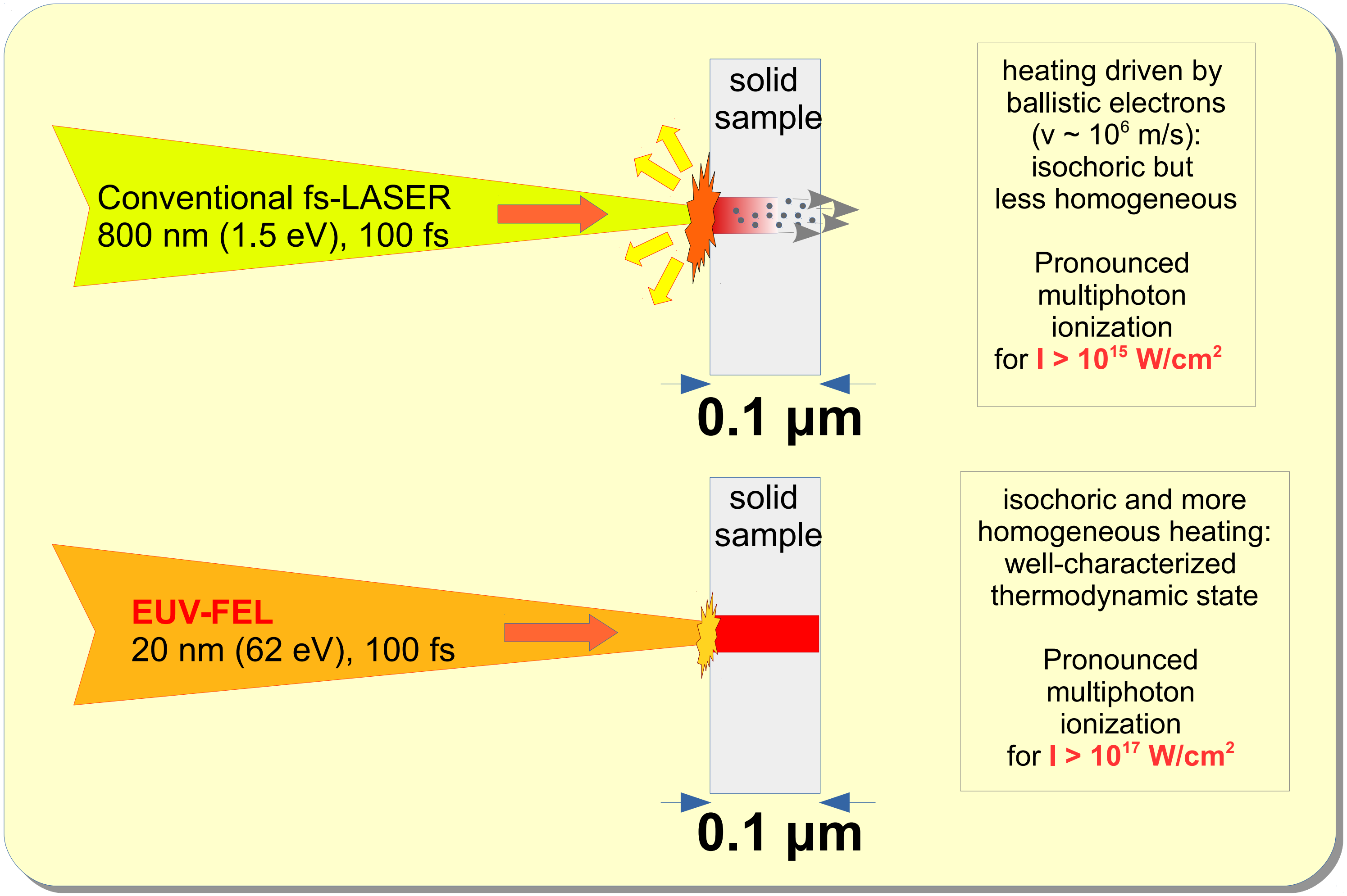
# Pulsed laser heating



# New accessible regions of the phase diag

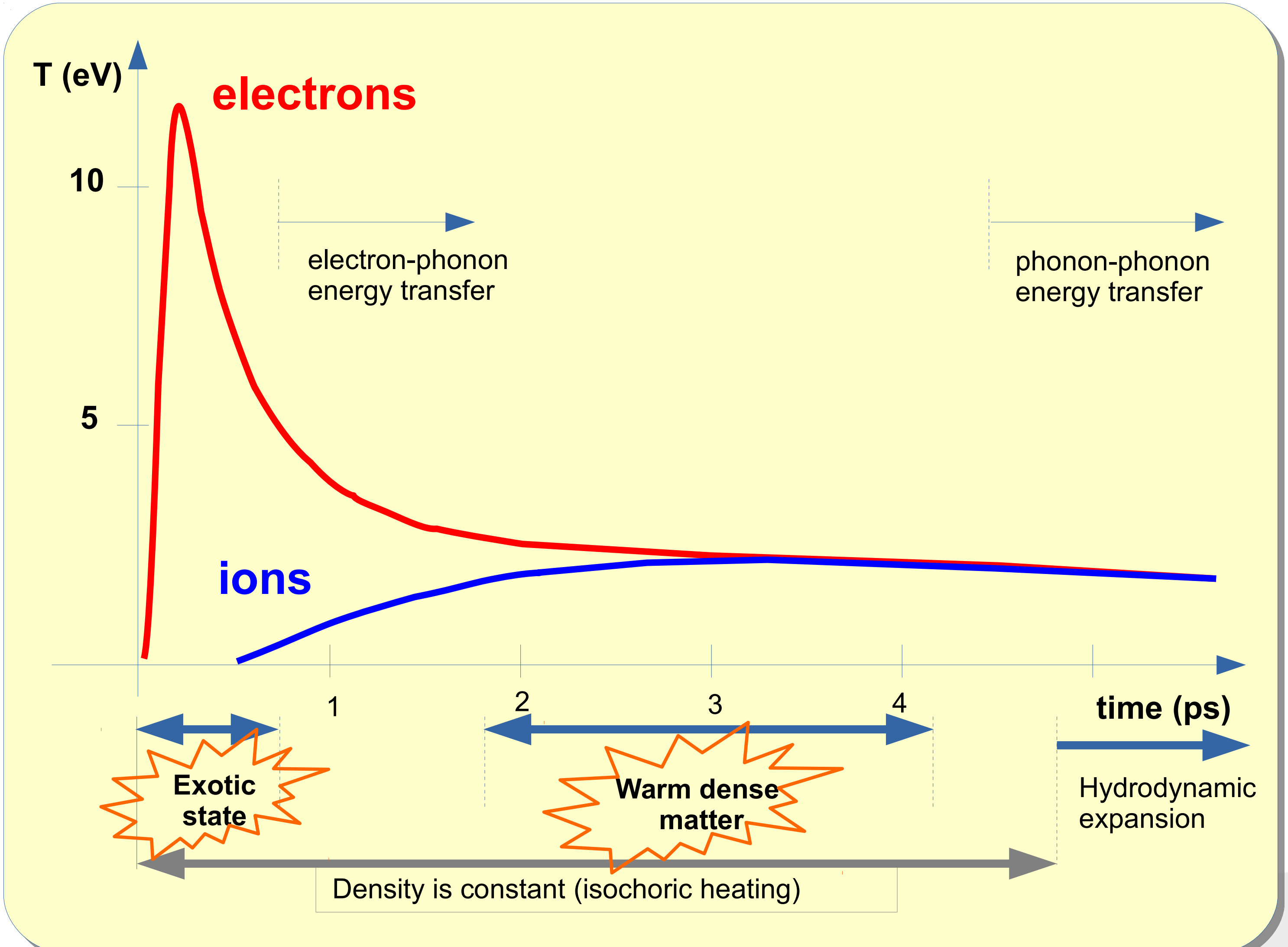
- Typically one knows the amount of energy delivered by the pump pulse and can calculate both the absorbed energy E and excited volume V. If one assumes that energy dissipation is negligible in the sub-100 fs time scale, the internal pressure resulting from isochoric heating can be estimated:
  - $p = E/V$ : pressure is an energy density
- Originally (exotic regime) only the electron system is involved in the energy absorption. In free electron metals, free-electrons can be described as a gas. The absorbed energy goes into electron kinetic energy (from equipartition theorem) and ionization:
  - $E/V = 3/2\, n_e k_B T_e + \sum n_i \Phi_i$ ($n_e$ electron density, $T_e$ electron temperature, $n_i$ atoms density in the ionization state "i", $\Phi_i$ ionization potential of ion stage "i")
- If ionization is negligible, one gets a simple estimation of the max average electron temperature in the exotic regime
  - $k_B T_e = 2/3\, p/n_e$

- Fluence = p d ( $10^{-4}$ J/cm$^2$ = 1 GPa nm )

# Two-temperature model (TTM)

When exposed to a sub-ps high energy density light pulse, matter exhibits a transient nonequilibrium regime described by the TTM

Important states of matter occurring upon isochoric are:
- Exotic state
- WDM state



# TTM analytical solution

electron heat capacity

electron-phonon coupling factor

heat conductivity

excitation term

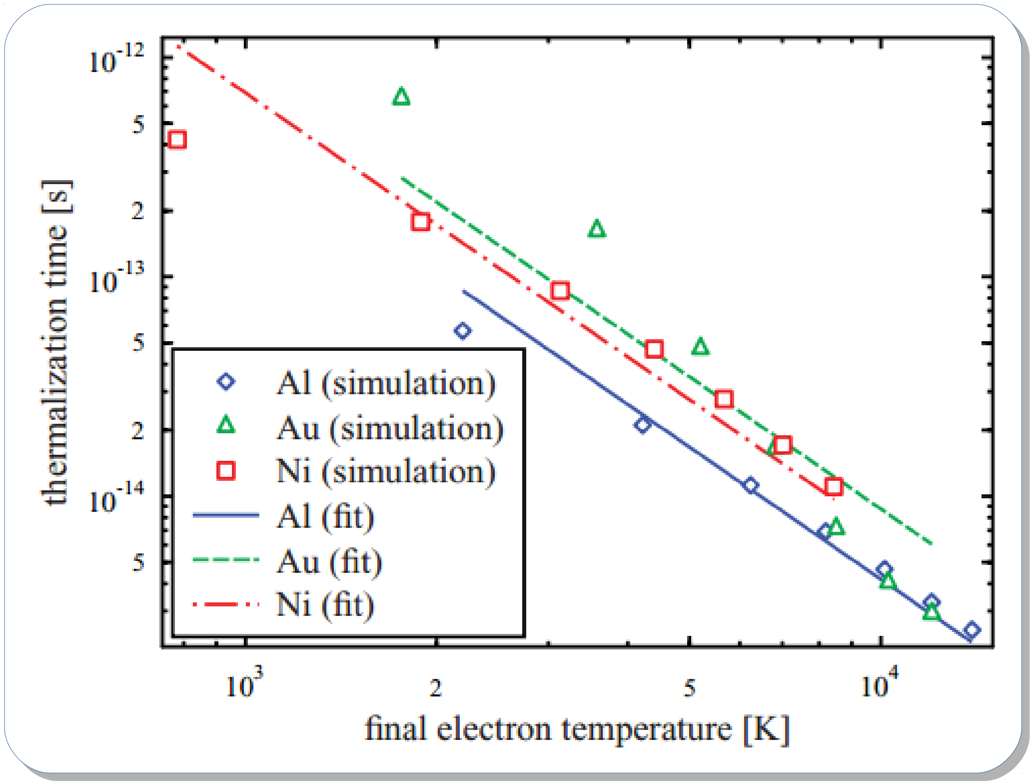
$$C_e(T_e) \cdot \frac{\partial T_e}{\partial t} = -\alpha \cdot (T_e - T_p) + \nabla(\chi_e \nabla T_e) + S(t),$$

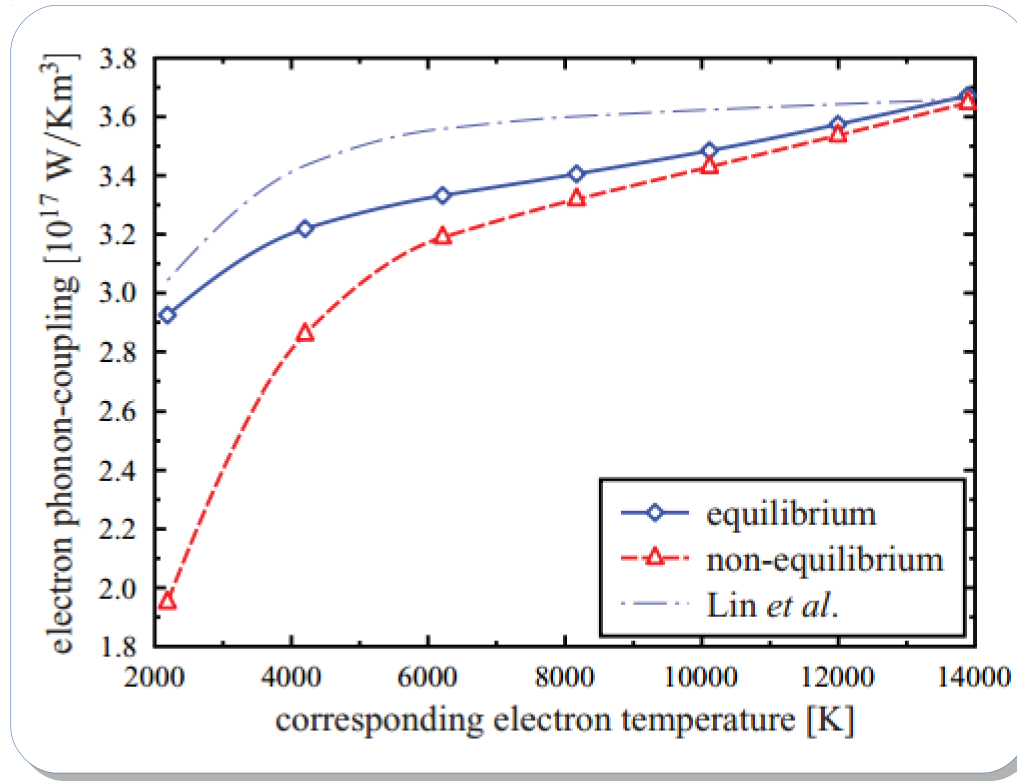
$$C_p(T_p) \cdot \frac{\partial T_p}{\partial t} = \alpha \cdot (T_e - T_p),$$

phonon heat capacity

Muller and Rethfeld, PRB 87 035139 (2013)

# Electron nonequilibrium properties

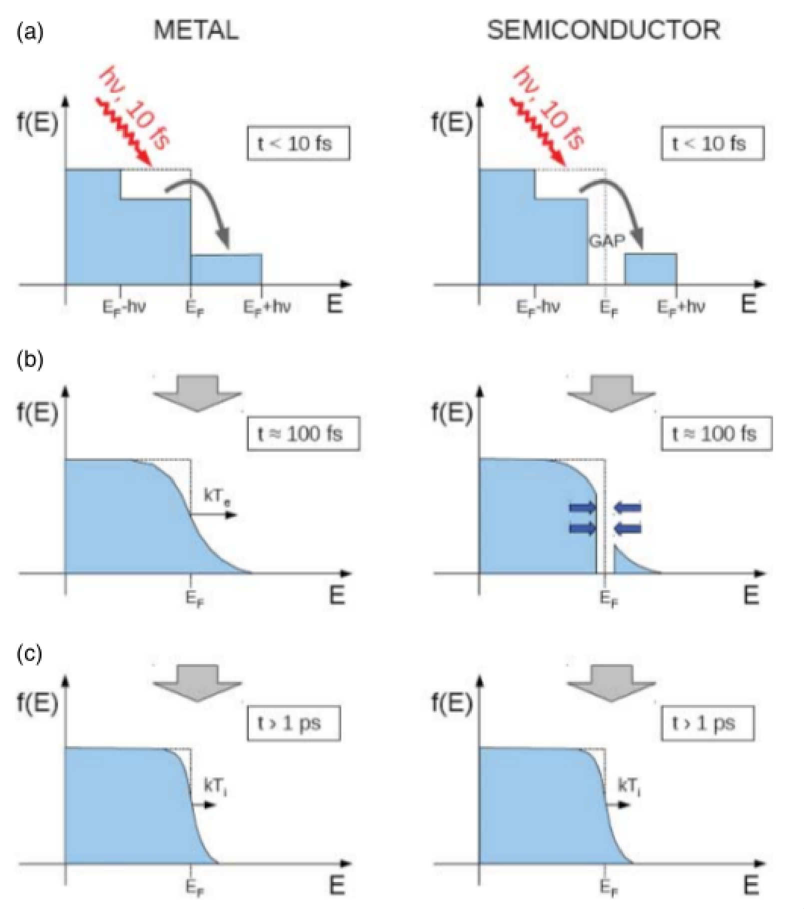


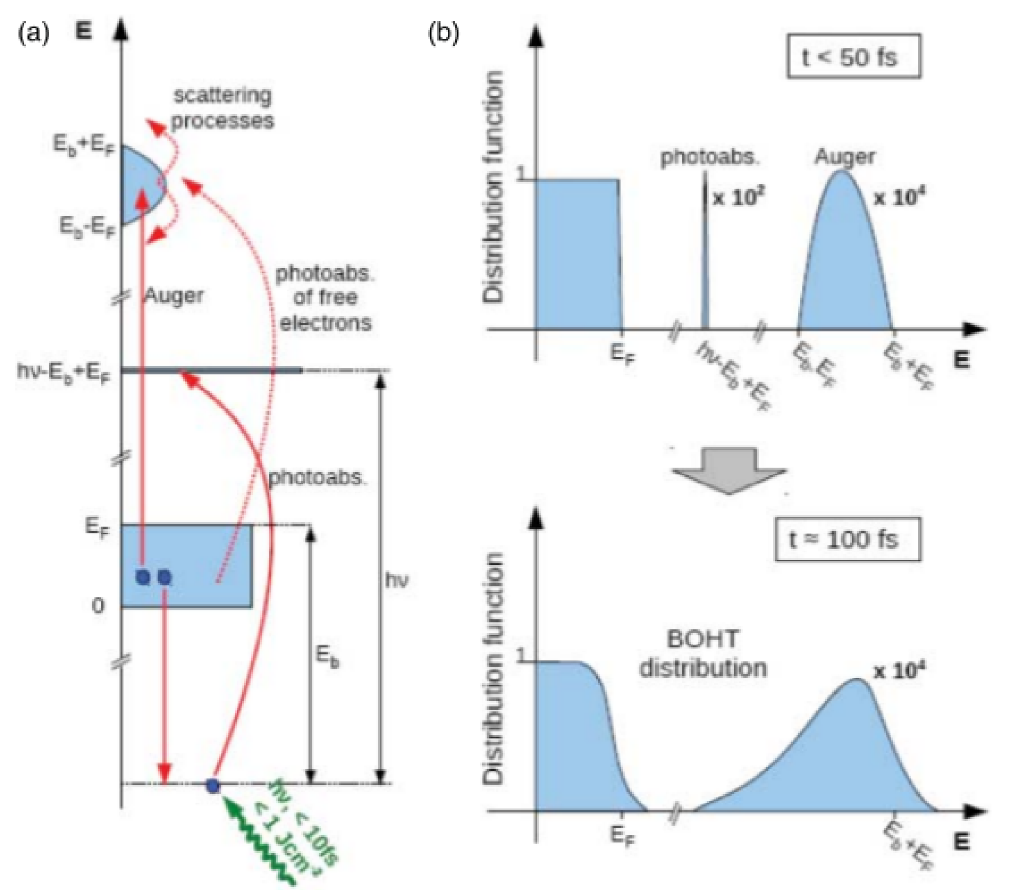

Muller and Rethfeld, PRB 87 035139 (2013)

# Ultrafast rearrangement of electron population (weak radiation field)

LASER-heating

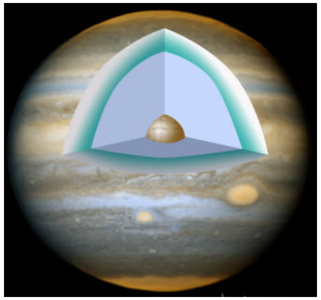


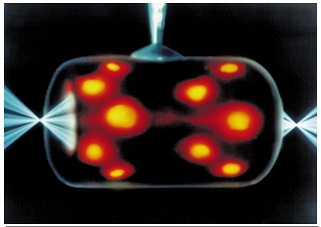

FEL-heating



# Warm dense matter (WDM) regime

- WDM can be found in:
  - cores of large planets
  - inertial confinement fusion
  - laser-induced plasma devices





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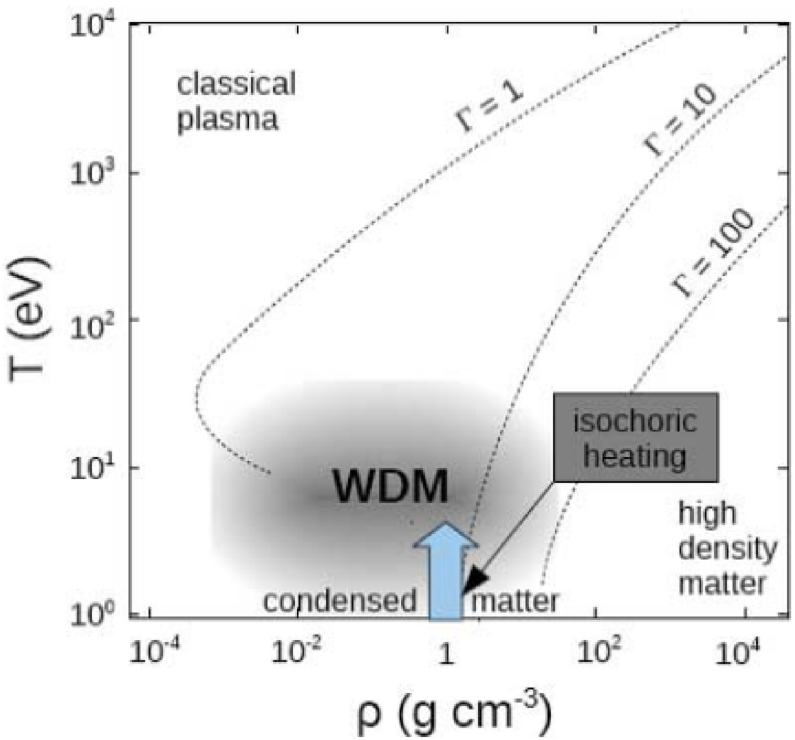
# Warm dense matter (WDM) regime




Coupling parameter

$$\Gamma = (Ze)^2 / (r_i k_B T_i)$$ Coulomb energy / kinetic energy

$$\theta = T_e / T_F$$ electron temperature / Fermi temperature

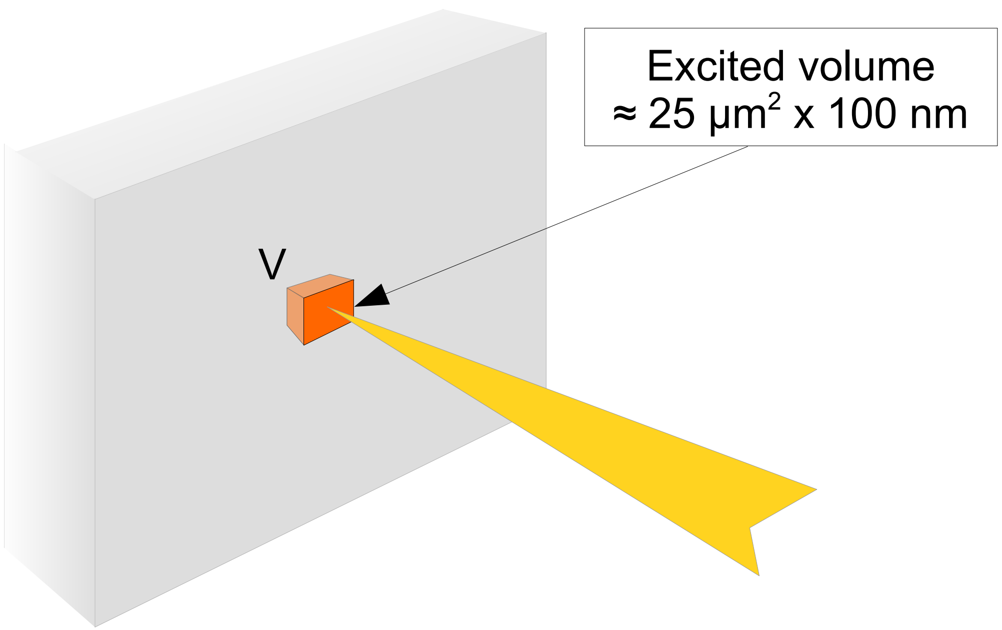
Degeneracy parameter

**Condensed Matter**: $\Gamma >> 1$ (high ion coupling), $\theta << 1$ (fully degenerate electron gas)

**WDM**: $\Gamma \geq 1$(strongly coupled ion plasma), $\theta \approx 1$ (partially degenerate electron gas)

**Classical plasma**: $\Gamma << 1$ (negligible ion coupling), $\theta >> 1$ (non-degenerate electron gas)

# Pressure conditions created by pulsed laser heating




**With free boundaries and after sufficient long time, the effect of temperature rise (at constant pressure) is thermal expansion ($\Delta V>0$).**
**With fixed boundaries, an effect of a temperature rise must be an internal (thermal) pressure**

**The max level of thermal pressure is significantly affected by the time regime of heating.**
**Sub-ps pulses can heat the excited volume prior to atomic structure expansion, thus generating enormous internal pressures.**

# Thermodynamics

$$C_p(T,p) = -T\left(\frac{\partial^2 G_m}{\partial T^2}\right)_p, \text{ molar heat capacity} \tag{1}$$

$$V_m(T,p) = \left(\frac{\partial G_m}{\partial P}\right)_T, \text{ molar volume} \tag{2}$$

$$\alpha_V(T,p) = \frac{1}{V_0}\left(\frac{\partial V_m}{\partial T}\right)_p, \text{ thermal expansion} \tag{3}$$

$$\kappa(T,p) = -\frac{1}{V_0}\left(\frac{\partial V_m}{\partial p}\right)_T, \text{ isothermal compressibility} \tag{4}$$

$$K = \kappa(T,0), \text{ isothermal compressibility at zero pressure} \tag{5}$$

$$V_m(T,p) = A\big(1 + nKp\big)^{-\frac{1}{n}} \exp\big(f(T)\big) \tag{6}$$

$$\Delta V_m(T_0,p) < 0, \text{ compression driven by external pressure rise at constant temperature}$$

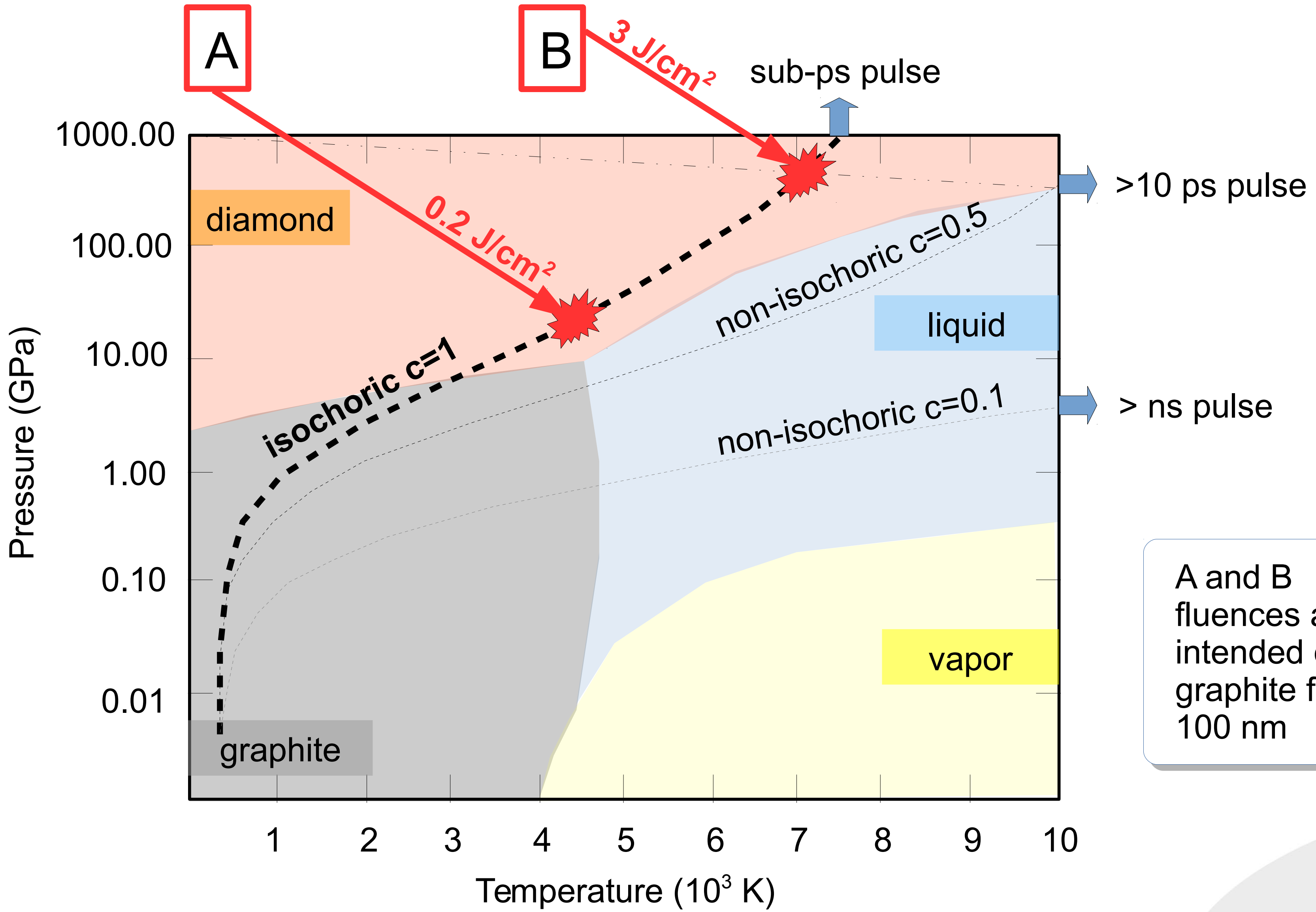
$$\Delta V_m(T,p_0) > 0, \text{ thermal expansion driven by temperature rise at constant pressure}$$

$$\frac{\Delta V_m(T_0,p)}{\Delta V_m(T,p_0)} = -c,\ 0 < c < 1 \tag{7}$$

$$\text{if } c = 1 \Rightarrow \Delta V_m(T,p_0) = \Delta V_m(T_0,p) \Rightarrow \text{isochoric heating}$$

$$p(T) = \frac{1}{nK}\left[\Big(1 - c\big(\exp(f(T) - f(T_0))\big)\Big)^{-n} - 1\right],\ p_0 \approx 0 \tag{8}$$

# Exploring the phase diagram of materials through pulsed laser-heating




A and B fluences are intended on a graphite foil of 100 nm

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