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# Excitation-induced non-thermal melting effects in silicon

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## Introduction

- We developped electron temperature-dependent interaction potentials for silicon to study the dynamics of silicon under laser irradiation.
- Electron temperature-dependent potentials give rise to electron temperature-dependent lattice expansion, elastic constants, and melting temperature.
- Novel material dynamics under laser irradiation can be observed with the new potential and electron-phonon coupling.
- Experimental ablation depth is reproduced with surprising accuracy.

## Interatomic potential

Functional form:

- Modified Tersoff potential (MOD):

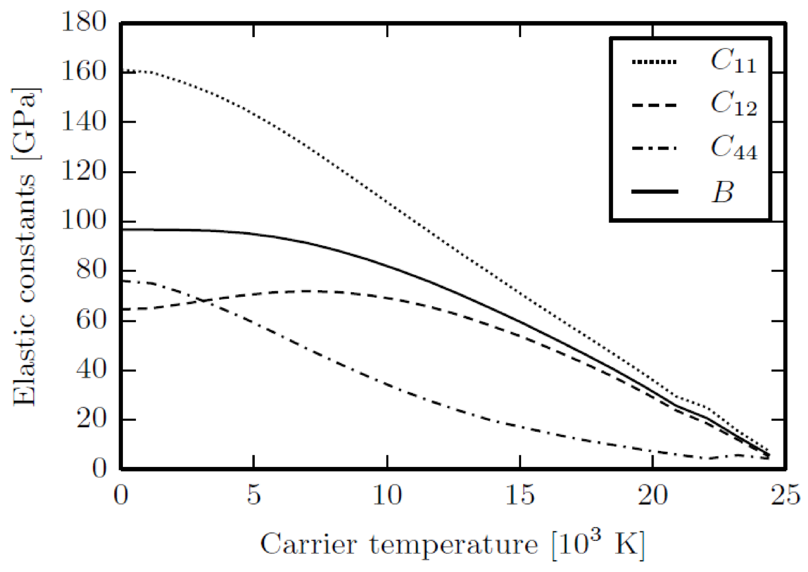
$$V = \frac{1}{2}\sum_i \sum_{j\neq i} f_C(r_{ij})\left[a_{ij} f_R(r_{ij}) + b_{ij} f_A(r_{ij})\right]$$

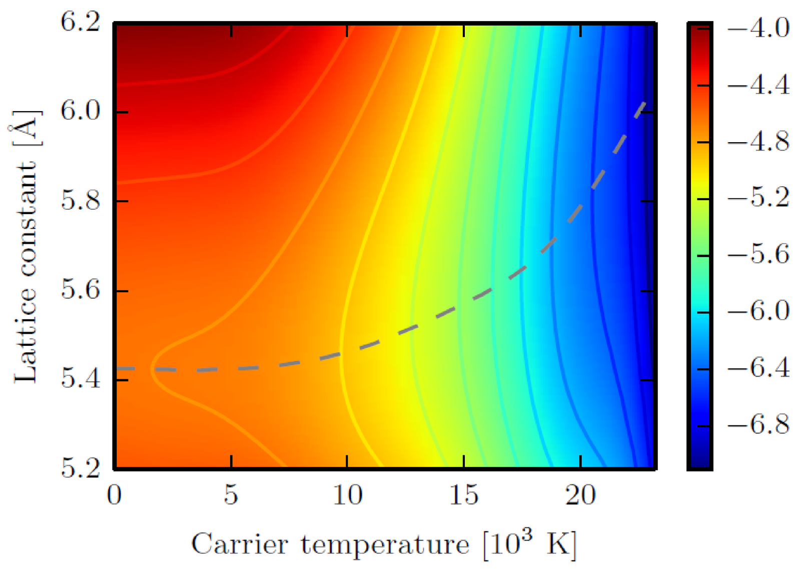
$$f_A(r_{ij}) = -B\exp(-\lambda_2 r_{ij}) \quad \text{and} \quad f_R(r_{ij}) = A\exp(-\lambda_1 r_{ij})$$

- Electron temperature-dependent parameters (MOD*) [1]:

$$P_t(T_c) = \sum_{n=0}^{m} a_n (k_B T_c)^n$$

$T_c$-dependent properties:




## $T_c$-dependent phase diagram

Thermodynamic integration:

- Hamiltonian of two systems coupled by $\lambda$:

$$H = H_1 + \lambda(H_2 - H_1).$$

- Work from switching $\lambda$ over time:

$$W^s_{1\to 2} = \int_{t_1}^{t_2}\left[\frac{\partial H(\lambda)}{\partial\lambda} + p\frac{\partial V(\lambda)}{\partial\lambda}\right]\frac{\partial\lambda}{\partial t}\,dt$$

⇒ Increase or decrease temperature over time at different constant pressures.
⇒ Dissipated energy $E_d$ vanishes if temperature is changed slowly.

- Calculation of Gibbs free energy:

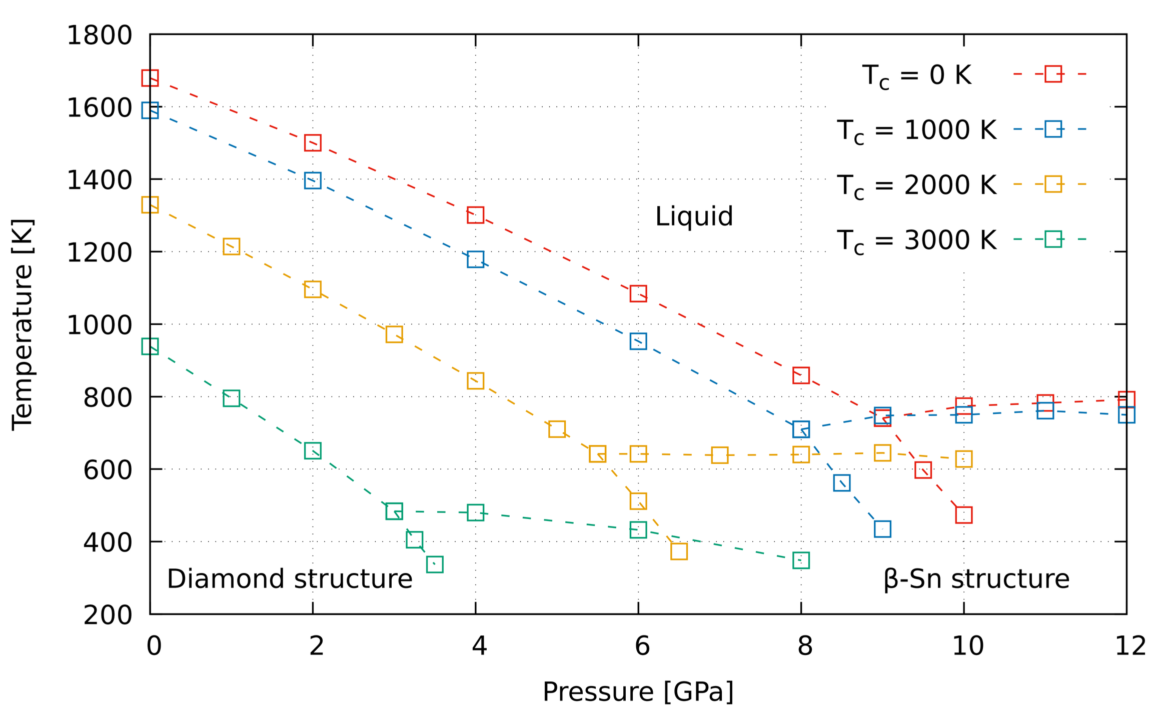
$$\Delta G = \frac{1}{2}\left(W^s_{1\to 2} - W^s_{2\to 1}\right)$$

$$\Rightarrow G(N,p,T_2) = G(N,p,T_1) - \frac{3}{2}k_B T_2 N \ln\left(\frac{T_2}{T_1}\right) + \frac{T_2}{T_1}\Delta G$$

Pressure-temperature phase diagram:

- Phase transitions are found from Gibbs free energy intersections.
- Phase transitions are shifted to lower temperatures at higher electron temperatures.

⇒ $T_c$-dependent potential is source of non-thermal effects.




## Simulation model

The Boltzmann equation of the electron distribution in the drift-diffusion limit yields the continuity equation of the charge carrier density and the energy

$$\frac{\partial n_c}{\partial t} = -\vec\nabla\cdot\vec J + \frac{dn_c}{dt} \quad \text{and} \quad \frac{\partial U}{\partial t} = -\vec\nabla\cdot\vec W + \frac{dU}{dt}$$

with the total carrier flux density

$$\vec J = -D_0\left(\vec\nabla n_c + \frac{2n_c}{k_B T_c}\vec\nabla E_g + \frac{n_c}{2T_c}\vec\nabla T_c\right)$$

and total energy flux

$$\vec W = (E_g + 3n_c k_B T_c)\,\vec J - \kappa_c\vec\nabla T_c.$$

Optical properties of the material are included via

$$\frac{dI}{dx} = -(\alpha_{SPA} + \alpha_{FCA})\,I - \beta_{TPA} I^2$$

and

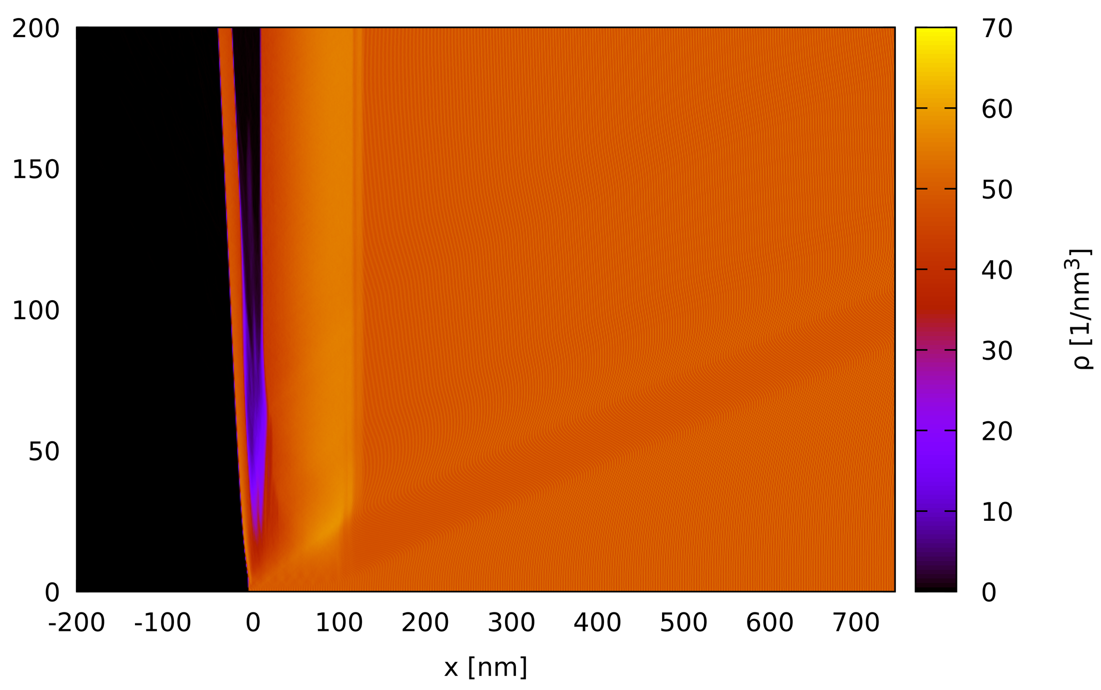
$$\frac{dn_c}{dt} = \sum_k I^k\frac{\alpha_k}{kh\nu} + \delta_{imp}n_c - \gamma_{Aug}n_c^3.$$

The atoms are moved via MD.

## Material dynamics during laser treatment

MOD:

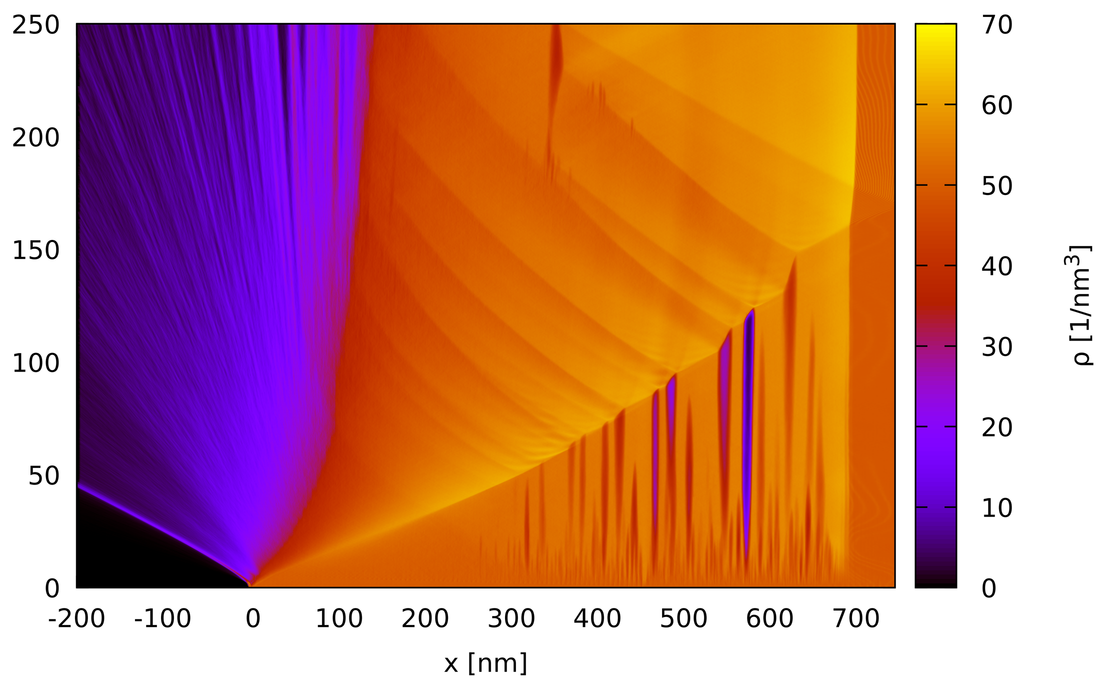
- Phase explosion (thermal)
- Skin layer forms detaches from the sample
- Pressure wave travels though material




MOD*:

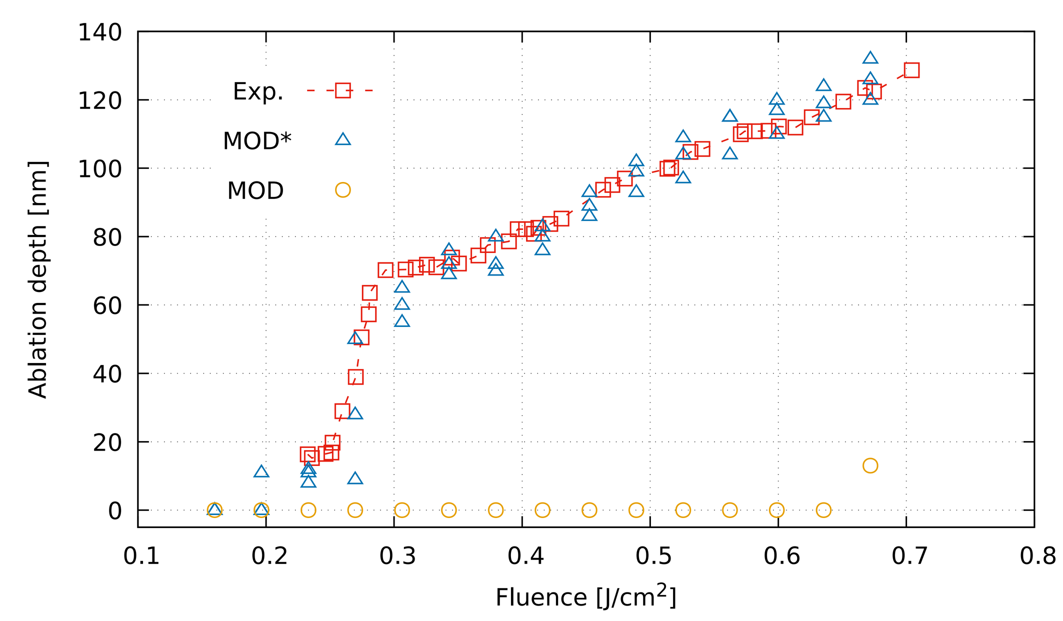
- Non-thermal surface evaporation
- Pre-shockwave melting
- Non-thermal void formation
- Void-induced liquid spallation

⇒ More complex non-thermal material dynamics




## Ablation depth from MOD and MOD*

- MOD doesn't reproduce correct ablation behaviour.
- MOD* reproduces experimental ablation depth accurately.
- Experimental data taken from [2].




**References:**

[1] Alexander Kiselev. "Molecular dynamics simulations of laser ablation in covalent materials". PhD thesis. 2017.

[2] Hao Zhang. "Single-shot femtosecond laser ablation on the nanoscale". PhD thesis. 2013.