The Atomistic-Continuum Modeling of Short Pulse Laser melting of Semiconductors

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Outline

1. Experiments of laser irradiation of silicon

2. Continuum approach to modeling silicon

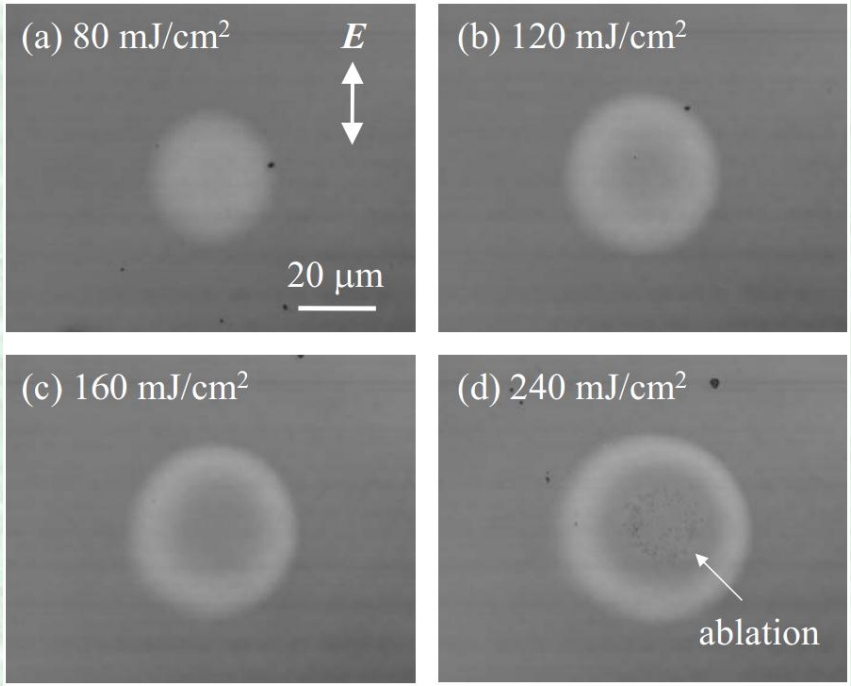
3. MD-TTM coupled approach

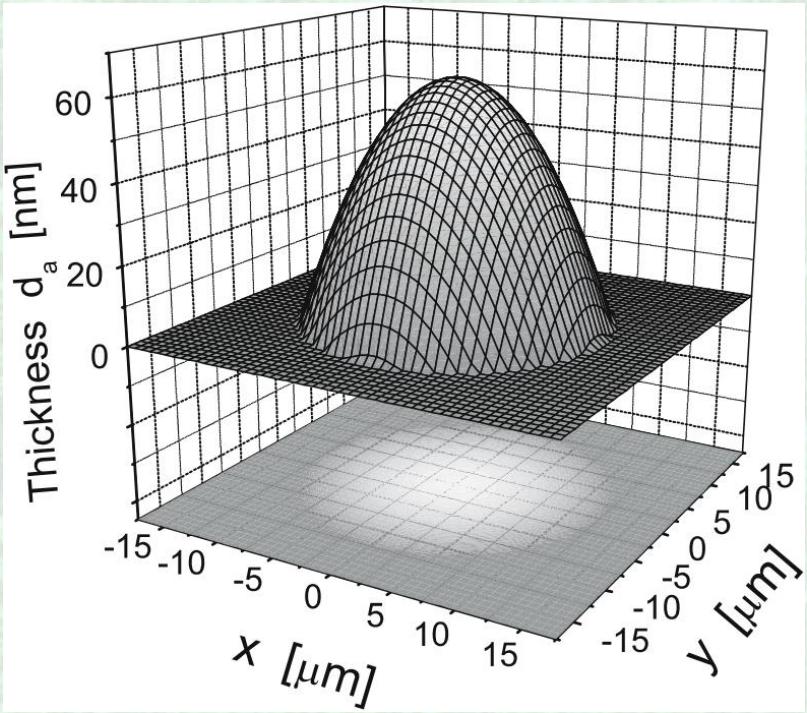
4. First results:
   - Continuum model vs. MD-TTM
   - Comparison to experiment

5. Conclusion

6. Future work

Motivation: nanostructuring on silicon





Processed Si surface by a single shot of 267 nm femtosecond laser irradiation. (LSM images)

Y. Izawa et al., J. Appl. Phys. 105, 064909 (2009)

3D-profile of the amorohous layer thickness induced by 130fs, 800nm laser pulse with fluence 4200 J/m²

J. Bonse, Appl. Phys. A: Mater. Sci. Proc., A 84, 63-66 (2006)

Model requirements

- **Absorption: generation of e--h+ pairs due to 1-photon and 2-photon absorption; absorption by free carriers**

- **Strong nonequilibrium state between laser-excited free carriers and lattice**

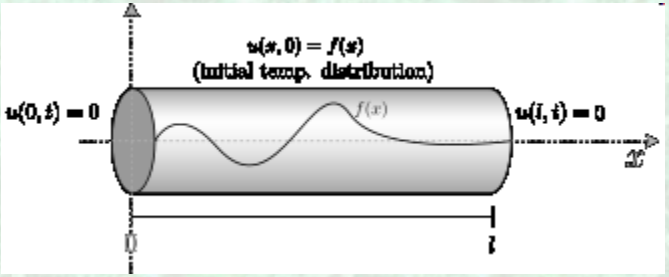
- **Fast heat conduction process due to free carriers**

- **Fast free carrier diffusion process**

- **Fast nonequilibrium phase transitions**

Part I

Continuum modeling of silicon (TTM)

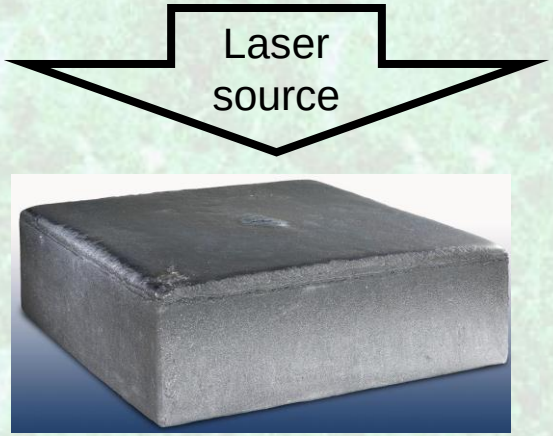


$$\frac{\partial T}{\partial t} = a\frac{\partial^2 T}{\partial x^2}$$

Continuum modeling for silicon

Based on: relaxation-time approximation of Boltzmann equation
(H.M. van Driel, Phys. Rev. B 35 8166 (1987))

Silicon : atoms and free carriers (electrons and holes)



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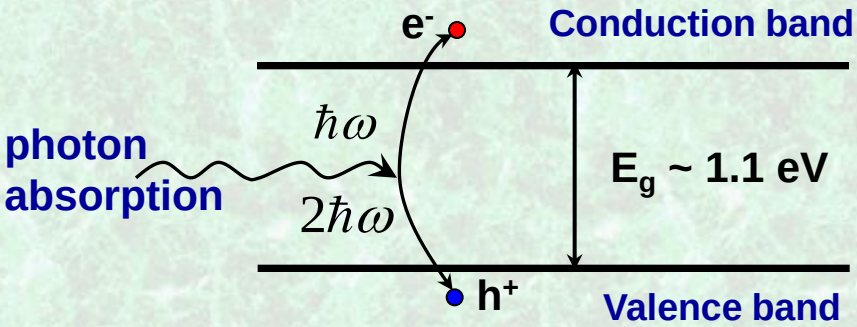


$$C_a \frac{\partial T_a}{\partial t} = div\left(k_a \nabla T_a\right) + G\left(T_e - T_a\right)$$

Diffusion equation for atoms

$$f_{e,h}(E) = \frac{1}{e^{\frac{E - E_{F_{e,h}}}{k_B T_{e,h}}} + 1}$$

Fermi-Dirac distributions
for electrons and holes

Continuum modeling for silicon

Energy conservations for carrier subsystem:

$$\frac{\partial U_e}{\partial t} = Source(\vec{r},t) - div\vec{W}(\vec{r},t) - G(T_e - T_L)$$

Energy of electrons in conduction band

Laser source

Energy flux

Energy exchange with atoms

$$Source(\vec{r},t) = \alpha I(\vec{r},t) + \beta I^2(\vec{r},t) + \Theta n I(\vec{r},t)$$

1-photon absorption

2-photon absorption

Free carriers absorption

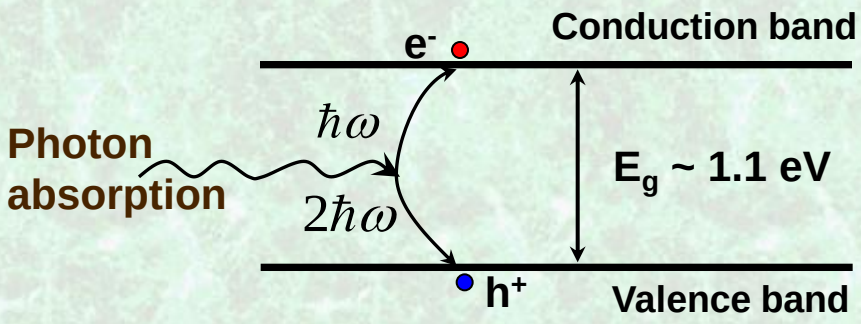
Continuum modeling for silicon

Carrier density and diffusion in conduction band

$$\frac{\partial n}{\partial t} =$$

Density of carriers in conduction band

Continuum modeling for silicon

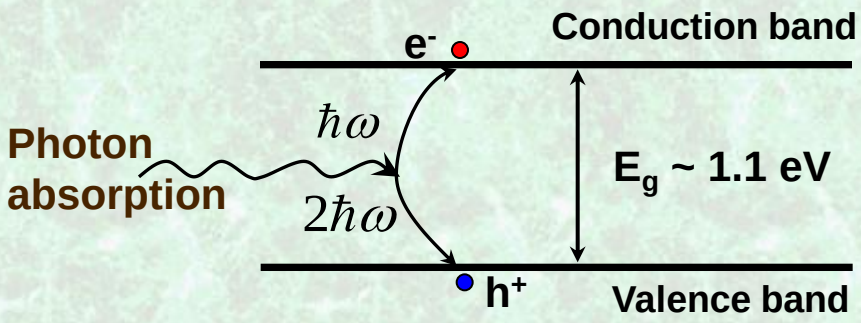


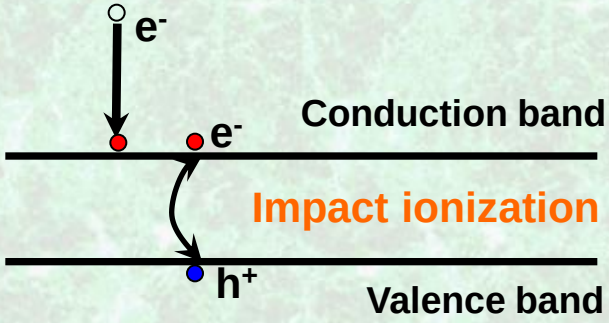
$$\frac{\partial n}{\partial t} = Source_n(\vec{r}, t)$$

Density of carriers in conduction band

Excitation of new carriers

Continuum modeling for silicon





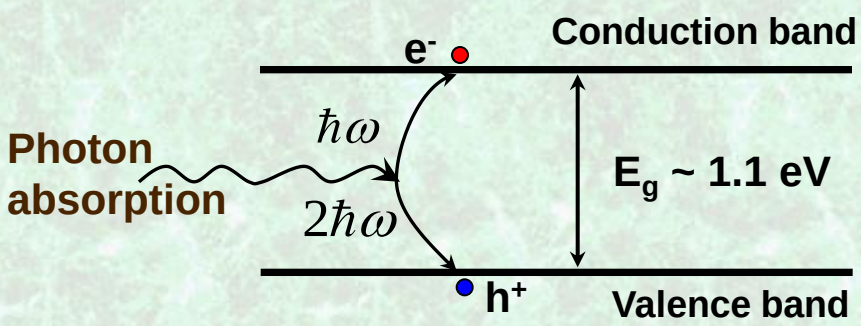
$$\frac{\partial n}{\partial t} = Source_n(\vec{r}, t) + \theta n$$

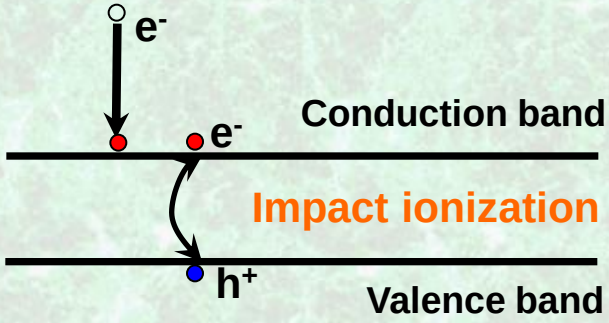
Density of carriers in conduction band

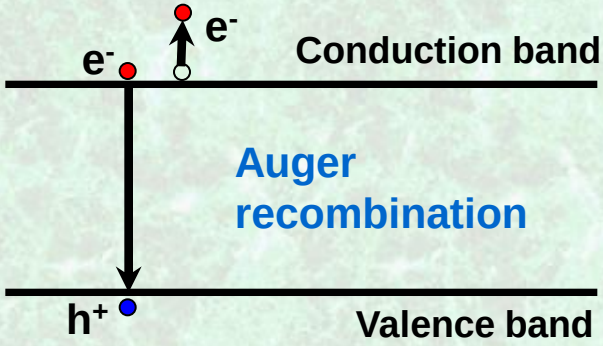
Excitation of new carriers

Impact ionization

Continuum modeling for silicon



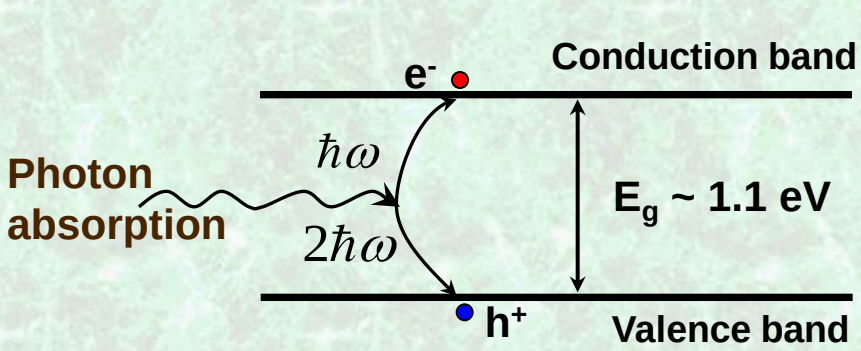


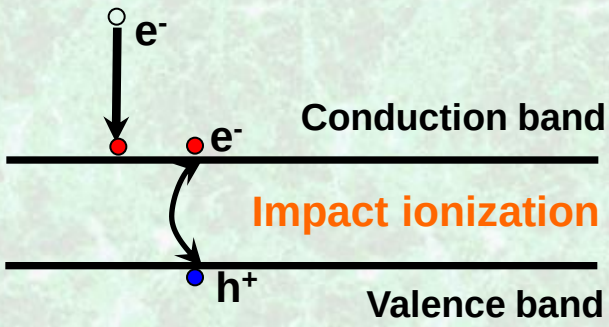


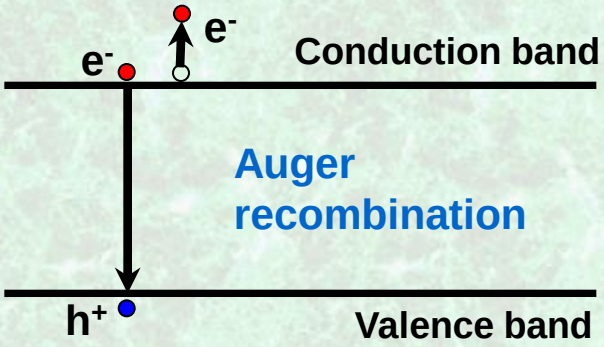
$$\frac{\partial n}{\partial t} = Source_n(\vec{r},t) + \theta n - \gamma n^3$$

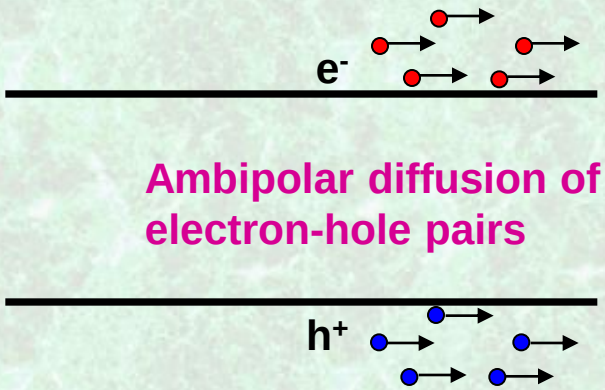
- Density of carriers in conduction band
- Excitation of new carriers
- Impact ionization
- Auger recombination

Continuum modeling for silicon









$$\frac{\partial n}{\partial t} = Source_n(\vec{r},t) + \theta n - \gamma n^3 - div(\vec{J})$$

Density of carriers in conduction band

Excitation of new carriers

Impact ionization

Auger recombination

Ambipolar diffusion

Continuum modeling for silicon

Self-consistent model on Si (analogue to TTM)

H.M. van Driel, Phys. Rev. B 35 8166 (1987)

$$\begin{cases} \dfrac{\partial n}{\partial t} = \dfrac{\alpha I}{\hbar\omega} + \dfrac{\beta I^2}{2\hbar\omega} + \theta n - \gamma n^3 - div\left(\vec{J}\right) \\ \dfrac{\partial U}{\partial t} = \left(\alpha + \Theta n\right) I + \beta I^2 - div\left(\vec{W}\right) - G\left(T_e - T_a\right) \\ C_a \dfrac{\partial T_a}{\partial t} = div\left(k_a \nabla T_a\right) + G\left(T_e - T_a\right) \end{cases}$$

Carrier density

Energy of excited carriers

Lattice temperature

Continuum modeling for silicon

Self-consistent model on Si (analogue to TTM)

H.M. van Driel, Phys. Rev. B 35 8166 (1987)

$$\begin{cases} \dfrac{\partial n}{\partial t} = \underline{\dfrac{\alpha I}{\hbar\omega} + \dfrac{\beta I^2}{2\hbar\omega}} + \theta n - \gamma n^3 - div\left(\vec{J}\right) \\ \dfrac{\partial U}{\partial t} = \underline{(\alpha + \Theta n) I + \beta I^2} - div\left(\vec{W}\right) - \underline{G(T_e - T_a)} \\ C_a \dfrac{\partial T_a}{\partial t} = div\left(k_a \nabla T_a\right) + \underline{G(T_e - T_a)} \end{cases}$$

Carrier density

Energy of excited carriers

Lattice temperature

Laser source ***Coupling term***

Solution algorithm for continuum part

$$\begin{cases} \dfrac{\partial U_e}{\partial t} = \underline{(\alpha + \Theta n) I + \beta I^2} - div\left(\vec{W}\right) - \underline{G\left(T_e - T_a\right)} \\ U = nE_{gap} + \dfrac{3}{2} n k_B T_e \left[H_{\frac{1}{2}}^{\frac{3}{2}}(\eta_e) + H_{\frac{1}{2}}^{\frac{3}{2}}(\eta_h) \right] \end{cases}$$

Energy of excited carriers

Connection between energy and temperature for Fermi-particles

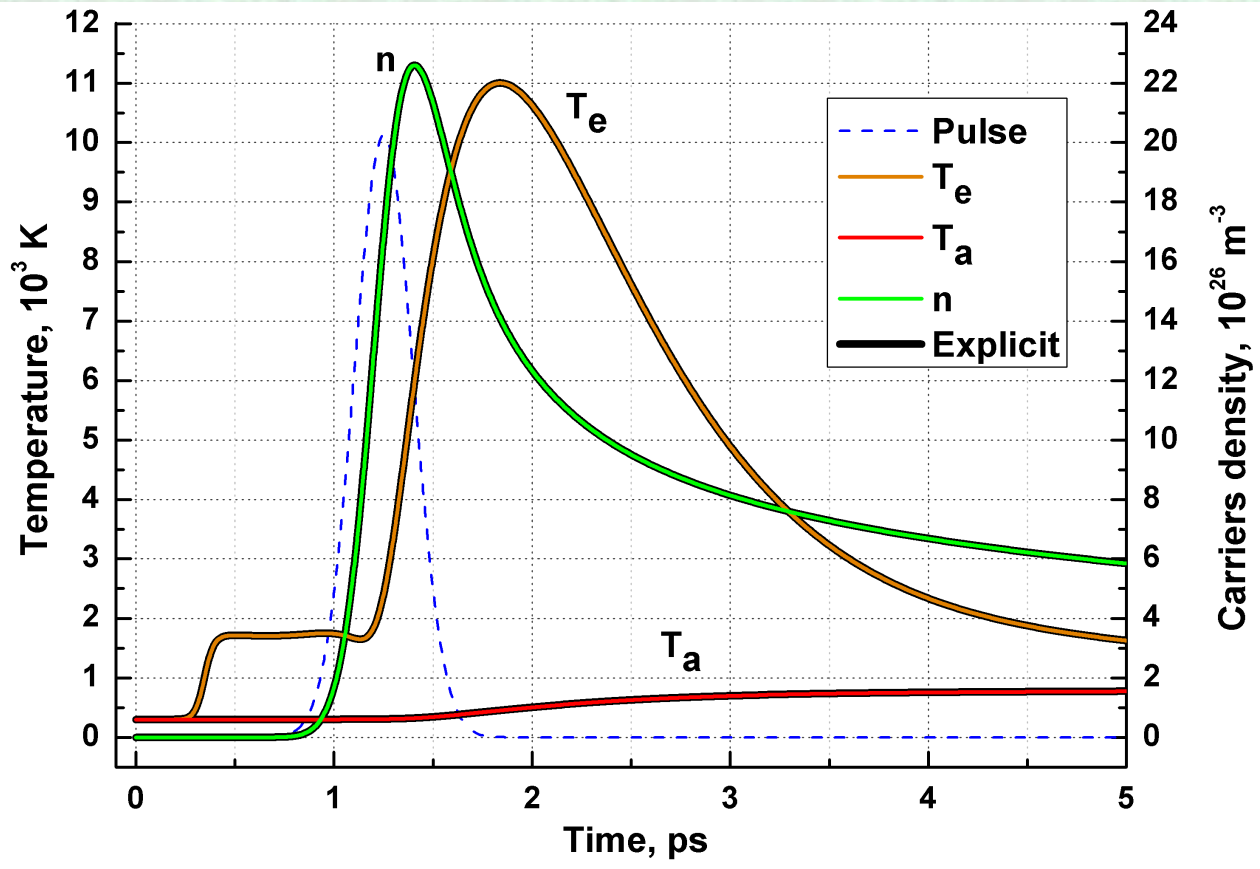
$$C_{e-h} \frac{\partial T_e}{\partial t} = \underline{(\alpha + \Theta n) I + \beta I^2} - div \vec{W}\left(T_e\right) - \underline{\frac{C_{e-h}}{\tau}\left(T_e - T_l\right)} + f\left(T_e, n\right)$$

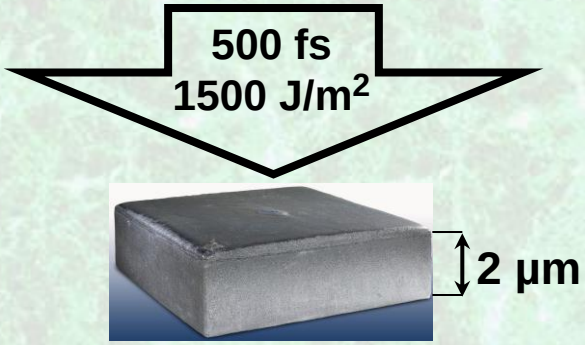
$$\frac{\partial T}{\partial t} = F\left(T, x, t, \frac{\partial T}{\partial x}, \frac{\partial^2 T}{\partial x^2} \right)$$

Diffusion equation. We use Crank-Nicolson half-implicit finite-differences method

Results of continuum calculations

Explicit and implicit schemes comparison





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- **First peak of T_e is due to small heat capacity of the carrier pairs**
- **Second peak of T_e is due to direct absorption by free carriers**

Calculation time 17 hours decreased to 23 seconds

Models for laser interaction with matter

Two-temperature model (TTM):

$$\begin{cases} \dfrac{\partial n}{\partial t} = \dfrac{\alpha I}{\hbar\omega} + \dfrac{\beta I^2}{2\hbar\omega} + \theta n - \gamma n^3 - \mathrm{div}\left(\vec{J}\right) \\ \dfrac{\partial U_e}{\partial t} = (\alpha + \Theta n) I + \beta I^2 - \mathrm{div}\left(\vec{W}\right) - G\left(T_e - T_a\right) \\ C_a \dfrac{\partial T_a}{\partial t} = \mathrm{div}\left(k_a \nabla T_a\right) + G\left(T_e - T_a\right) \end{cases}$$

+ accounts for electron-phonon nonequilibrium and fast electron heat conduction

– fails to describe kinetics of fast phase transition processes

Molecular dynamics (MD) method:

$$m_i \frac{d^2 \vec{r}_i}{dt^2} = \vec{F}_i = -\sum_{j \neq i} \vec{\nabla}_{\vec{r}_j} U\left(\vec{r}_i - \vec{r}_j\right); i = 1, \ldots, N$$

+ can describe kinetics of fast nonequilibrium phase transition processes

– the classical MD method does not have free electrons included explicitly

Combine the advantages of different approaches in a single model

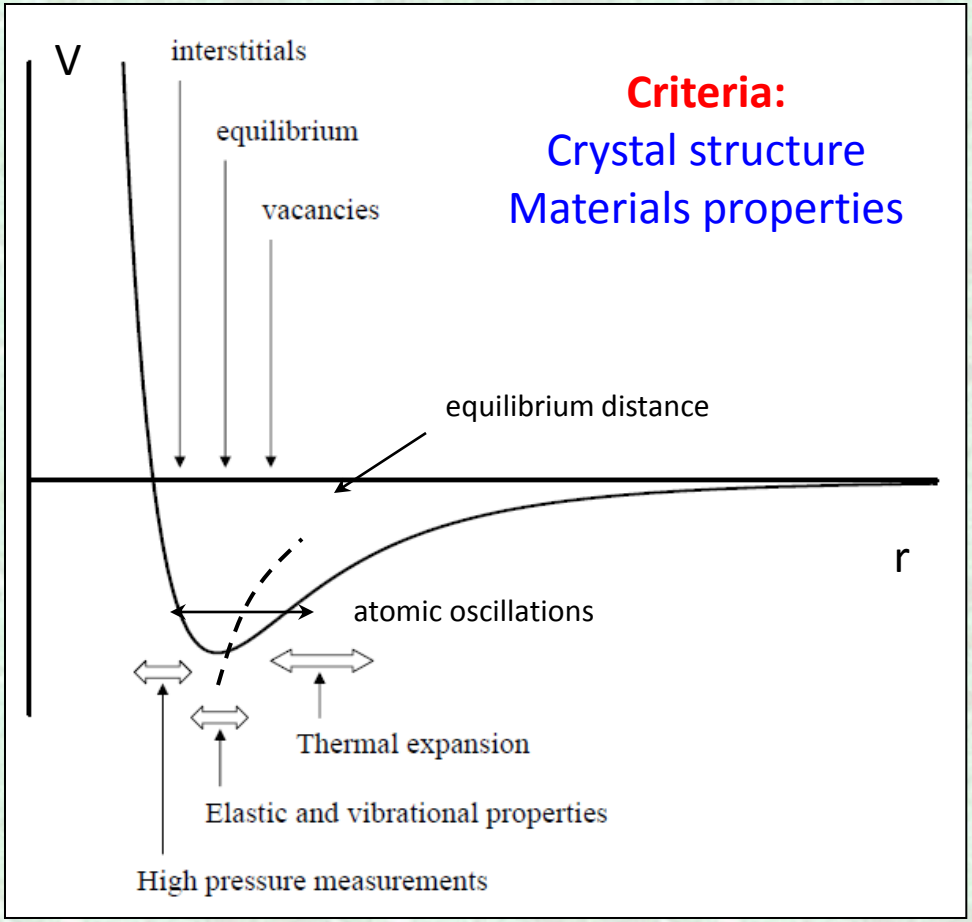
Choice of the potential of MD method

Molecular Dynamics:

$$m_i \vec{a}_i = \vec{F}_i = -grad\, V(\vec{r}_1, \vec{r}_2, \ldots \vec{r}_n)$$

Example: Lennard-Johns potential for Wan der Vaals interactions in inert gases Ar, Kr and molecular systems:

$$V = \sum_{i \neq j} U(r_{ij}) = \sum_{i \neq j} 4\varepsilon \left(\left(\frac{r_{ij}}{\sigma} \right)^{12} - \left(\frac{r_{ij}}{\sigma} \right)^{6} \right)$$

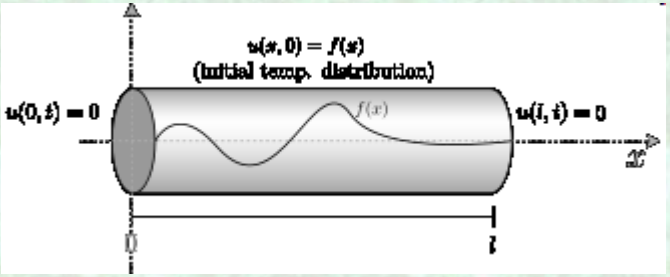


Stillinger-Webber potential with many body angular part for open diamond structures as in Si:

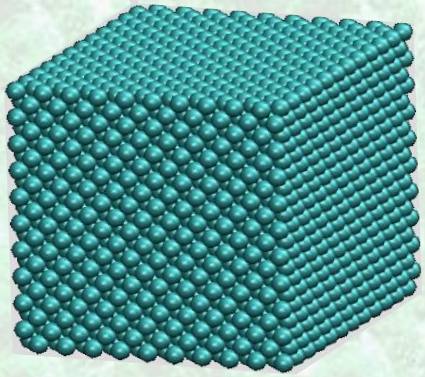
$$V = \frac{1}{2} \sum_{ij} U(r_{ij}) + \sum_{ijk} g(r_{ij}) g(r_{jk}) \left[\cos\theta_{ijk} + \frac{1}{3} \right]^2$$

Part II

Atomistic-continuum modeling of silicon (MD-TTM)



+



The atomistic-continuum model for silicon

$$\frac{\partial n}{\partial t} = \frac{\alpha I}{\hbar\omega} + \frac{\beta I^2}{2\hbar\omega} + \theta n - \gamma n^3 - div\left(\vec{J}\right)$$

carrier density

$$\frac{\partial U_e}{\partial t} = \left(\alpha + \Theta n\right) I + \beta I^2 - div\left(\vec{W}\right) - G\left(T_e - T_a\right)$$

electron-hole temperature

$$C_a \frac{\partial T_a}{\partial t} = div\left(k_L \nabla T_L\right) + G\left(T_e - T_a\right)$$

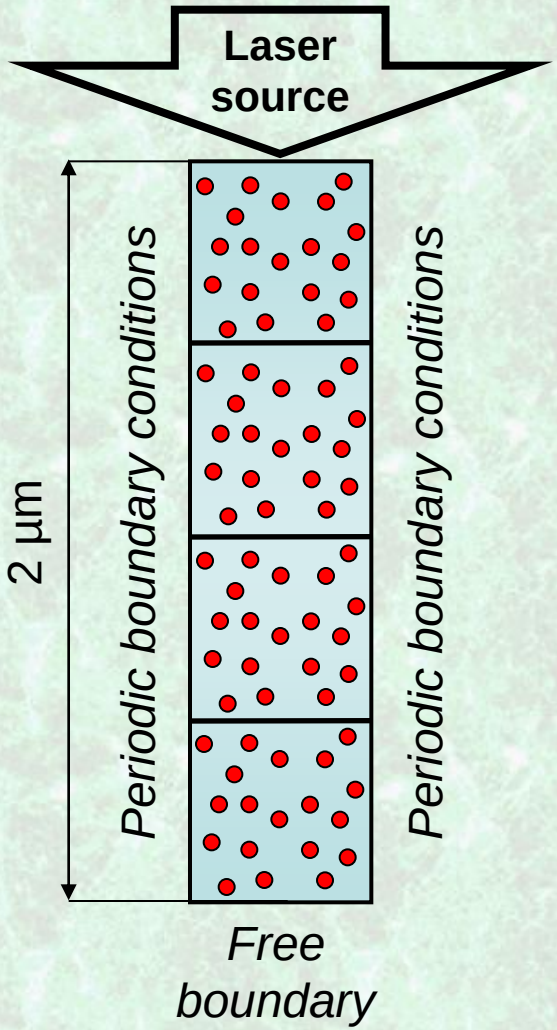
lattice temperature

Molecular dynamics for silicon atoms: (Stillinger-Weber potential)

$$m_i \frac{d^2 \vec{r}_i}{dt^2} = \vec{F}_i + \xi m_i \frac{d\vec{r}_i^{\,T}}{dt}$$

The atomistic-continuum model for silicon

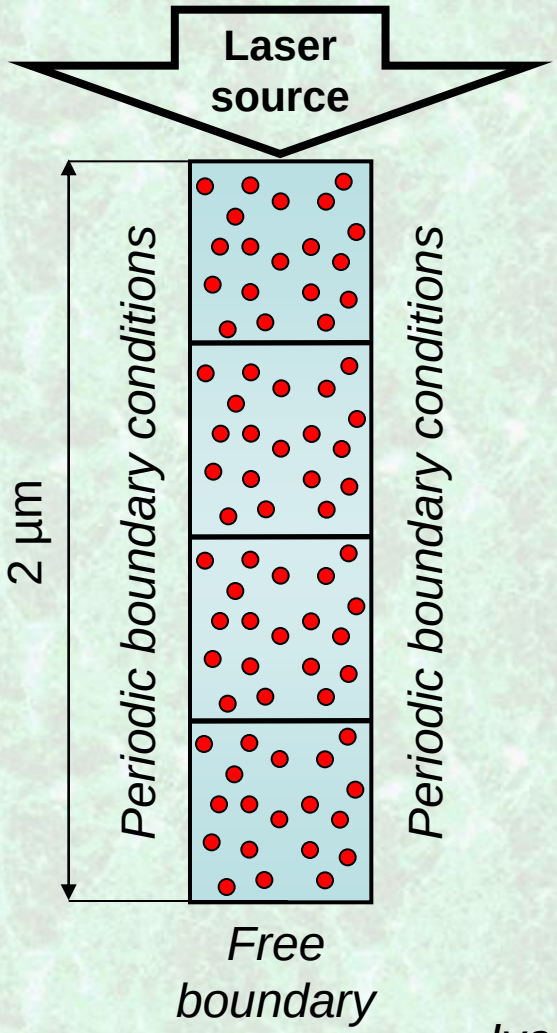
Coupling organization



$$G(T_e - T_a)$$

The atomistic-continuum model for silicon

Coupling organization



$$G(T_e - T_a)$$

Temperature of atoms:

a→e

$$\langle E_{kin} \rangle = \sum_{i=1}^{N_{atoms}} \frac{m_i \left(v_i^T\right)^2}{2} = \frac{3}{2} kT$$

"Friction" term for Molecular Dynamics:

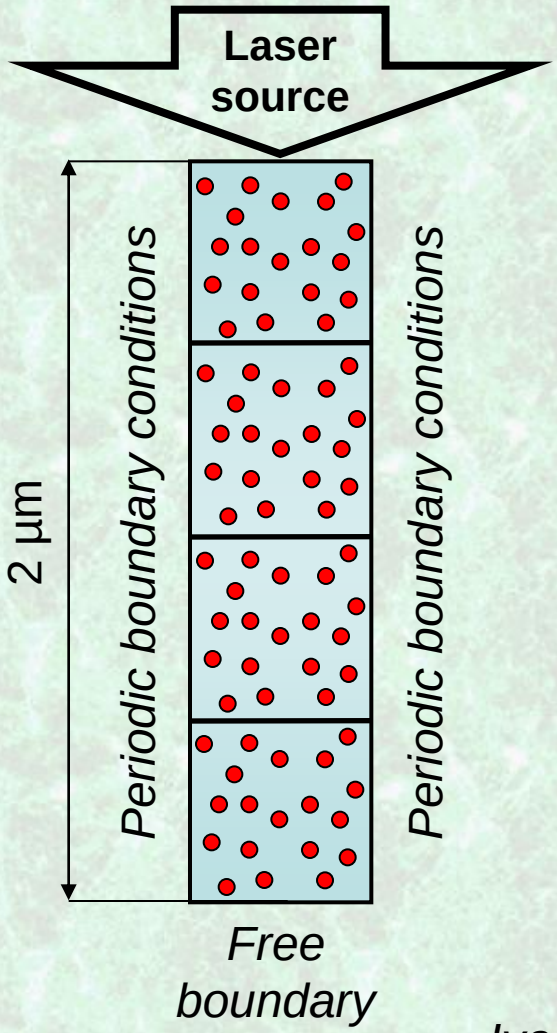
e→a

$$m_i \frac{d^2 \vec{r}_i}{dt^2} = \vec{F}_i + \xi m_i \frac{d\vec{r}_i^{\,T}}{dt}$$

Ivanov, Zhigilei, Phys. Rev. B 68, 064114 (2003)

The atomistic-continuum model for silicon

Coupling organization



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"Friction" term for Molecular Dynamics:

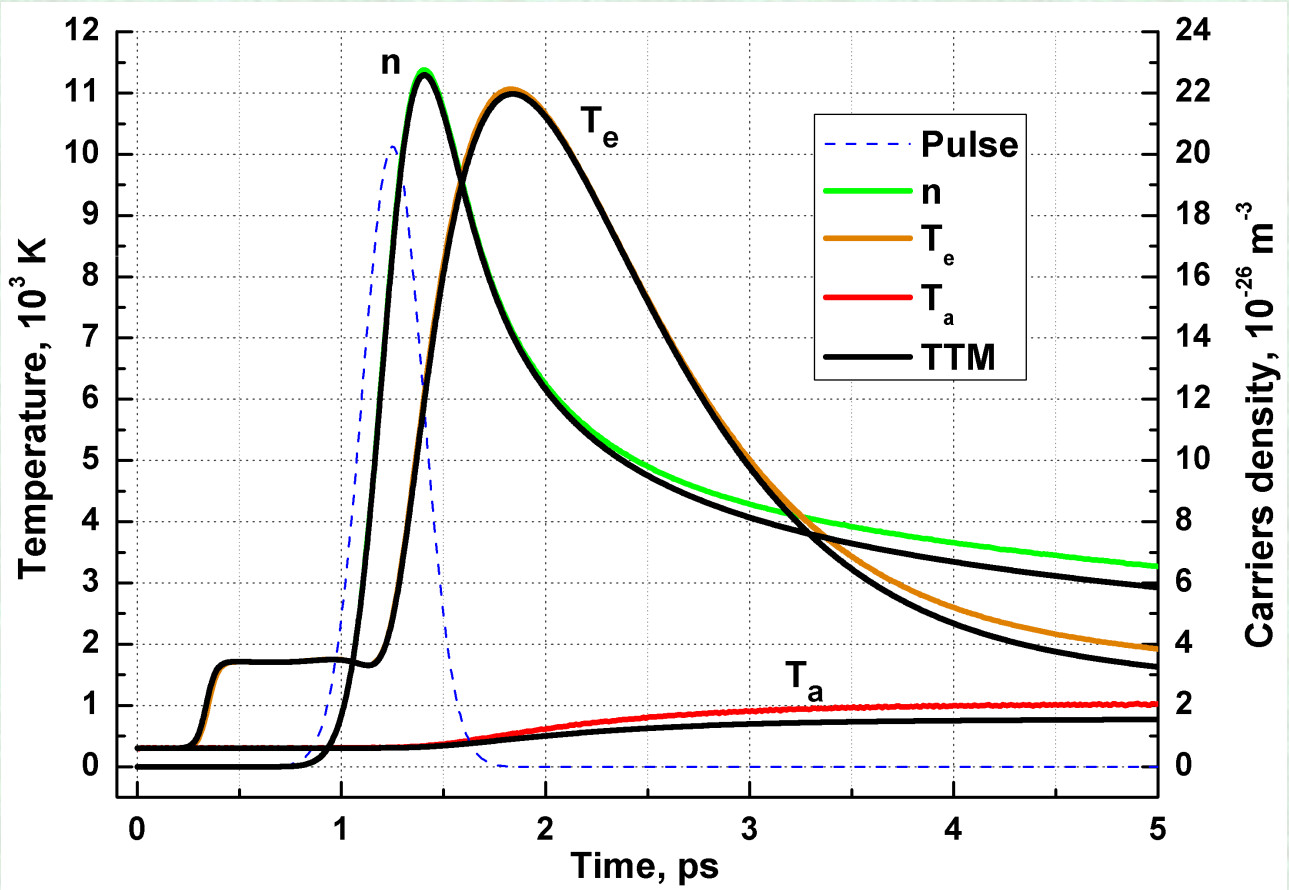
e→a

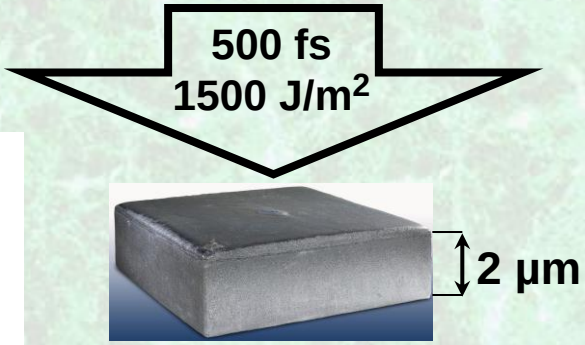
$$m_i \frac{d^2 \vec{r}_i}{dt^2} = \vec{F}_i + \frac{G V_N (T_e - T_a)}{2K^T} m_i \frac{d\vec{r}_i^{\,T}}{dt}$$

Ivanov, Zhigilei, Phys. Rev. B 68, 064114 (2003)

Results of MD-TTM calculations

MD-TTM compared to pure TTM





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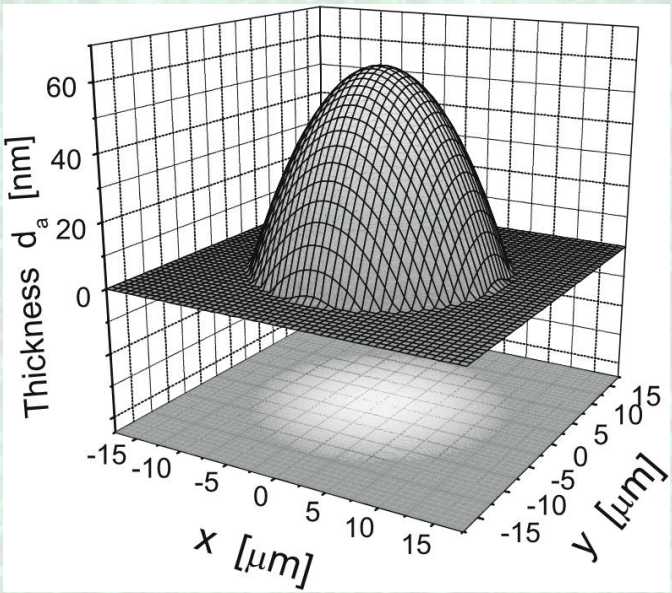
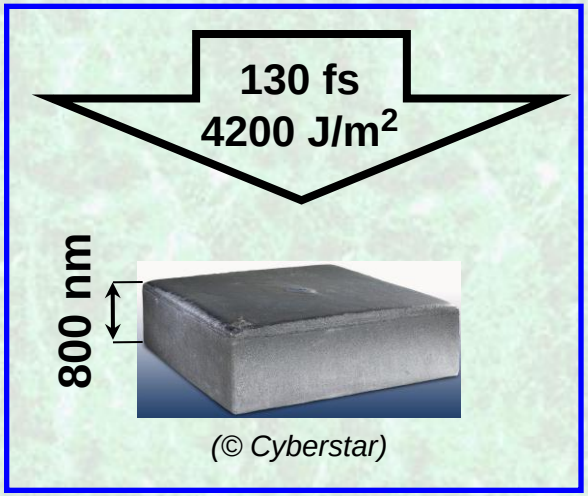
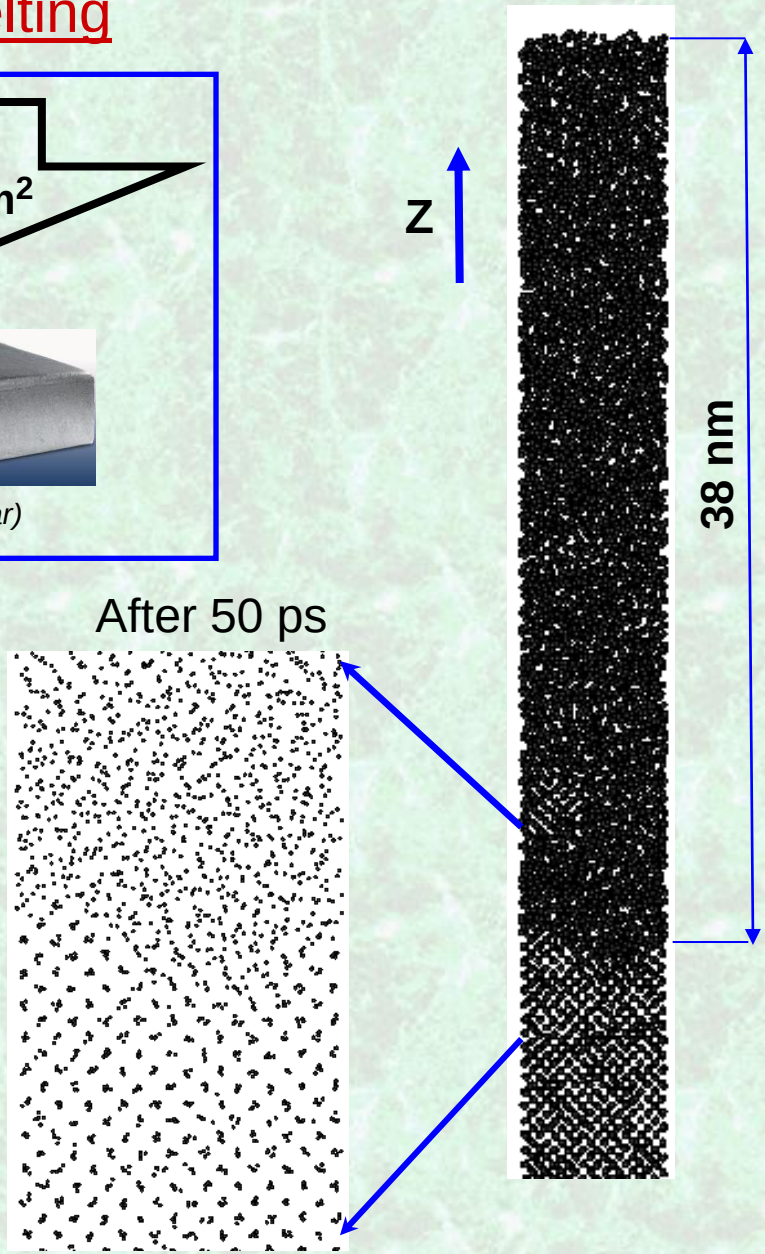
- **Difference in T_a due to surface effects: expansion of the sample and surface energy**
- **No difference for undersurface cells**

Results of MD-TTM calculations

Laser melting







Amorohous layer thickness induced by 130fs, 800nm laser pulse with fluence 4200 J/m²

J. Bonse, Appl. Phys. A: Mater. Sci. Proc., A 84, 63-66 (2006)

Conclusion

- Time step of continuum model has been increased by 10^4 times
- The atomistic-continuum model for laser interaction with Si has been implemented
- First calculations show the correctness of the new model
- First calculation of laser melting shows good qualitative agreement with experiment

Future work

- Utilization of the modified potential* accounting for the changes in atomic bonding versus parameters of photoexcited carriers
- Implementation of electron diffusion in 3D
- Realization of parallel algorithm
- Direct comparison of the model with an experiment

Shokeen, Schelling, J. Appl. Phys. 109, 073503 (2011)

Acknowledgements

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DFG

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Thank you for your attention!