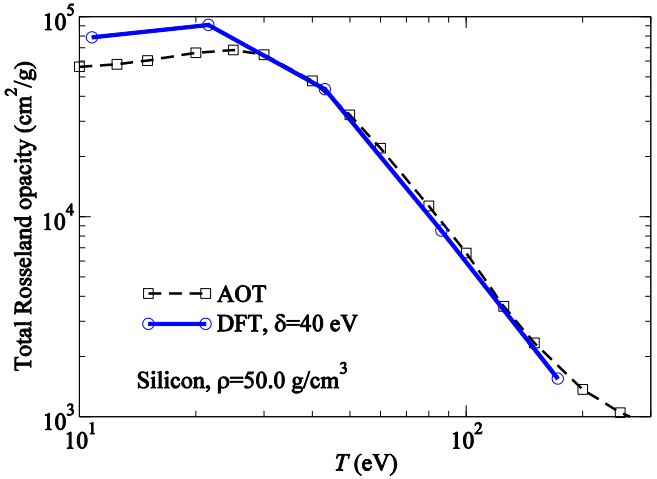
Systematic *Ab-initio* Calculations of Optical Properties of Silicon for Inertial Confinement Fusion Applications



AOT data are not accurate at low temperatures relevant to the ICF implosion conditions

V.V. Karasiev
University of Rochester
Laboratory for Laser Energetics

61st Annual Meeting of the American Physical Society
Division of Plasma Physics
Fort Lauderdale, FL
21-25 October 2019

Summary

Systematic first-principles calculations of absorption coefficients and opacities of silicon have been performed in a wide range of material densities and temperatures

- Our density-functional theory (DFT)-based calculations naturally account for quantum effects, continuum lowering, and Fermi-surface rising in hot dense silicon plasmas

- A set of accurate all-electron pseudo-potentials to calculate x-ray absorption near edge structure (XANES) spectra has been constructed for first time

- Absorption coefficients and opacities of silicon (densities between 0.1 and 500 g/cm^3 and T between 0.5 and 1000 eV) have been calculated

Collaborators

S. X. Hu

LLE, University of Rochester

L. Calderin

University of Arizona

Funding Acknowledgments:

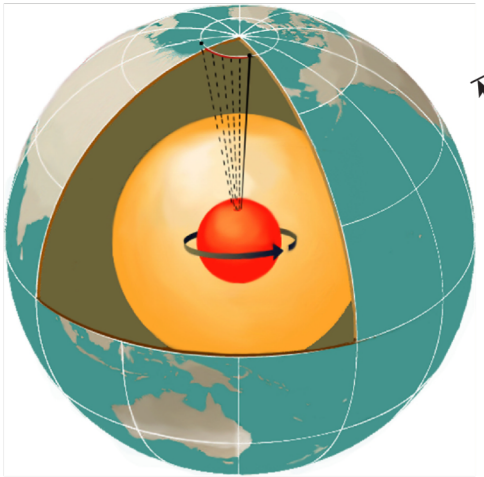
This work was partially supported by the U.S. National Science Foundation PHY Grant No. 1802964

This material is based upon work supported by the Department of Energy National Nuclear Security Administration under Award Number DE-NA0003856

Silicon is important to HED physics, such as planetary science and ICF capsules

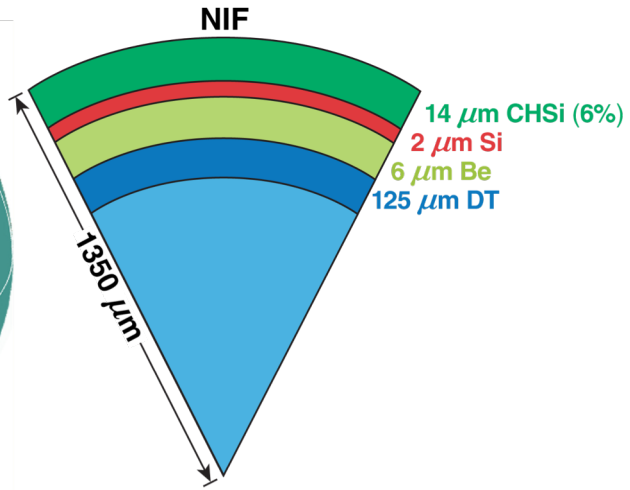
- The high-pressure silicon equation of state (EOS) is crucial to understanding the dynamics of silicon rich planets (i.e., Earth)
- Silicon is used in ICF capsules to reduce fuel preheat and laser–plasma instability (LPI) effects

Planetary science*



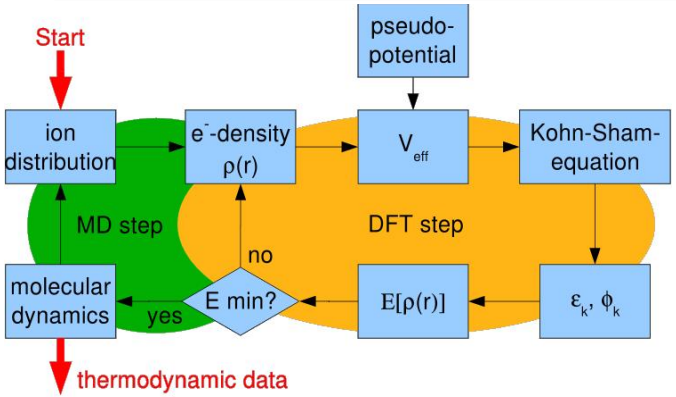
E25548d

ICF capsule**



* http://www.nasa.gov/sites/default/files/images/607068main_world-unlabeled.jpg
** V. N. Goncharov *et al.*, Phys. Plasmas 21, 056315 (2014).
HED: high-energy density
ICF: inertial confinement fusion
NIF: National Ignition Facility

Thermal density functional theory (DFT) coupled with *ab-initio* molecular dynamics (AIMD) is a powerful tool to study the properties of matter at extreme conditions



Ions: Classical treatment via Molecular dynamics

$$m_I \ddot{\mathbf{R}}_I = -\vec{\nabla}_I V(\mathbf{R}_1, \mathbf{R}_2, \ldots, \mathbf{R}_N)$$

Born-Oppenheimer energy surface:

$$V\left(\{\mathbf{R}\}\right) = \Omega\left(\{\mathbf{R}\}\right) + E_{\text{ion-ion}}\left(\{\mathbf{R}\}\right)$$

$\Omega[n] = \mathcal{F}[n] + \int d\mathbf{r}\ (v_{\text{ext}}(\mathbf{r}) - \mu)\, n(\mathbf{r})$ -- Grand potential

$\mathcal{F}[n] = \mathcal{F}_s[n] + \mathcal{F}_H[n] + \mathcal{F}_{xc}[n]$ -- Free-energy functional

$\mathcal{F}_H[n]$ -- Hartree energy

$\mathcal{F}_{xc}[n]$ -- Exchange-correlation (XC) free energy

$\mathcal{F}_s[n]$ -- Noninteracting (Kohn-Sham) free energy

Electrons: Quantum treatment via free-energy density functional theory, requires solution of Kohn-Sham system of coupled 0ne-electron equations.

Electronic transport coefficients are calculated from *ab-initio* simulations within the framework of Kubo-Greenwood approach (linear response theory)

UR LLE

The real part of electrical conductivity σ_1 and thermal conductivity κ are given in terms of the Onsager coefficients L_{mn}:

$$\sigma = \sigma_1 + i\sigma_2; \quad \sigma_1 = L_{11}; \quad \kappa = \frac{1}{T}\left(L_{22} - \frac{L_{12}^2}{L_{11}}\right)$$

The imaginary part σ_2 can be calculated via Kubo-Greenwood formula in terms of the velocity operator matrix elements or alternatively from the principal value integral.

The dielectric function and index of refraction (Re and Im parts, *n* and *k*):

$$\varepsilon(\omega) = \varepsilon_1(\omega) + i\varepsilon_2(\omega) = \left[n(\omega) + ik(\omega)\right]^2$$

$$\varepsilon_1(\omega) = 1 - \frac{4\pi}{\omega}\sigma_2(\omega); \quad \varepsilon_2(\omega) = \frac{4\pi}{\omega}\sigma_1(\omega)$$

$$n(\omega) = \sqrt{\frac{|\varepsilon(\omega)| + \varepsilon_1(\omega)}{2}}; \quad k(\omega) = \sqrt{\frac{|\varepsilon(\omega)| - \varepsilon_1(\omega)}{2}}$$

The reflectivity and absorption coefficient:

$$r(\omega) = \frac{\left[1 - n(\omega)\right]^2 + k^2(\omega)}{\left[1 + n(\omega)\right]^2 + k^2(\omega)}$$

$$\alpha(\omega) = \frac{4\pi}{n(\omega)c}\sigma_1(\omega)$$

Grouped Rosseland mean opacity:

$$K_R(\omega_1 : \omega_2) = \frac{\int_{\omega_1}^{\omega_2} n^2(\omega)\frac{\partial B(\omega, T)}{\partial T}\,\mathrm{d}\omega}{\int_{\omega_1}^{\omega 2} n^2(\omega)\frac{1}{\alpha_m(\omega)}\frac{\partial B(\omega, T)}{\partial T}\,\mathrm{d}\omega}$$

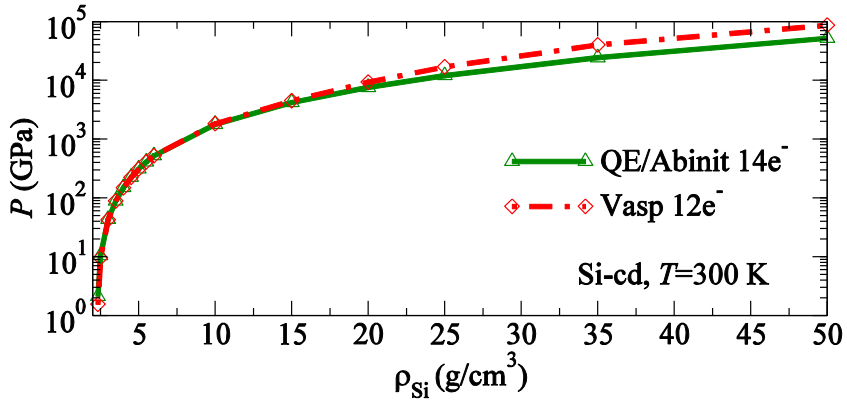
Calderin, Karasiev, Trickey, Comp. Phys. Commun. 221, 118 (2017)

Standard out-of-the-box DFT tools/software frequently fail at extreme conditions: There is a need to develop pseudopotentials transferable to high *P* and high *T*

• Calculation of optical properties within the x-ray range involves transitions from core states → explicit treatment of 1s electrons in the electronic structure calculations is required.

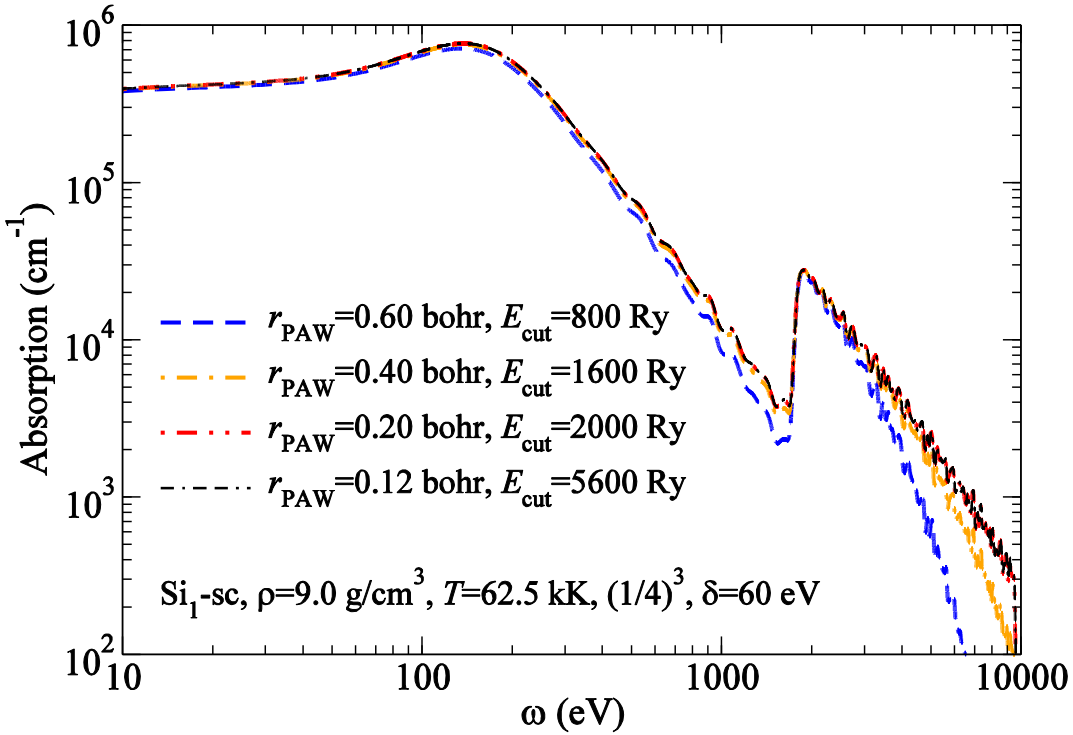
• Transferable all-electron PAW for Si, r_{PAW}=0.20 bohr has been constructed: V. V. Karasiev, S.X. Hu, V. Recoules, Comp. Phys. Commun. (in preparation);

• Standard Vasp 12-electron PAW fails at ρ > 10 g/cm^3

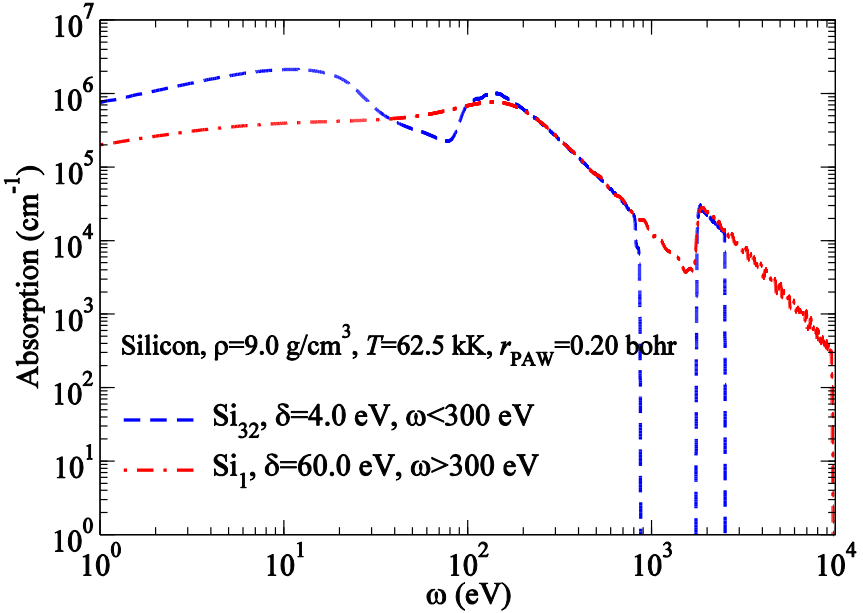


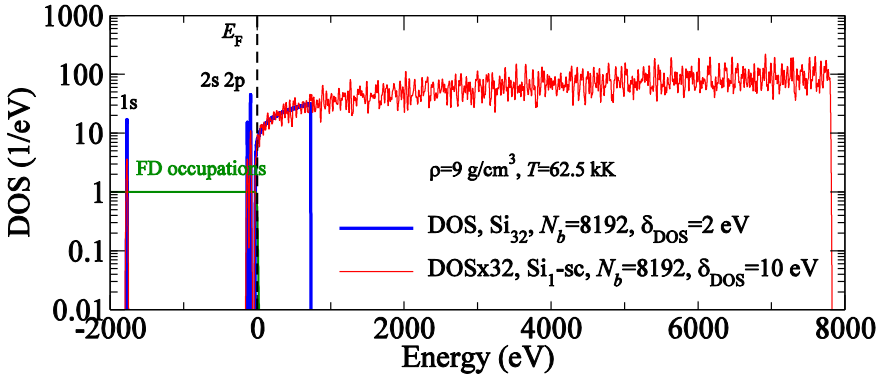
← Relative pressure error of VASP calculation reaches 60% at ρ > 35 g/cm^3

A convergence study of absorption w/r PP cutoff radius shows that a hard PP (small cutoff-radius) is required to converge the absorption coefficients at high photon frequencies

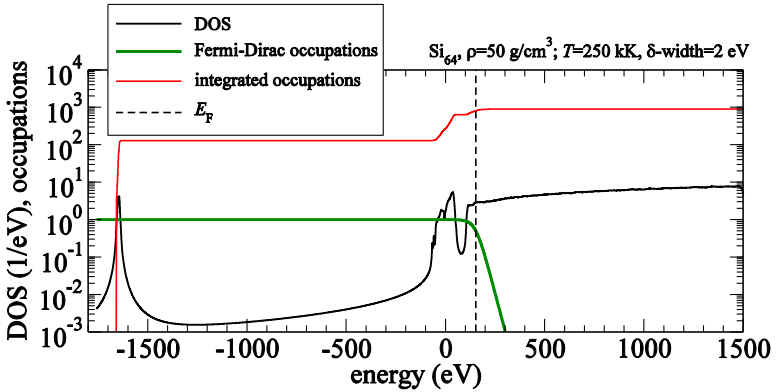


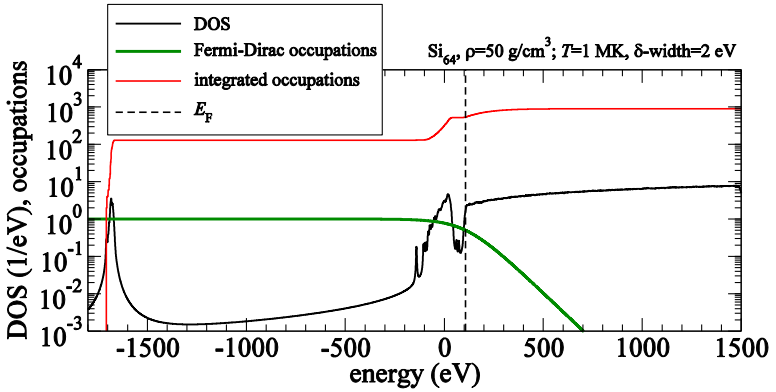
A novel method of extending calculations to the x-ray range was proposed. We combine the molecular dynamics (MD) snapshot (Si_{xx}) absorption data (for low-ω and near L-, K-edges) and periodic sc-Si_1 data (for the L- and K-edge tails)

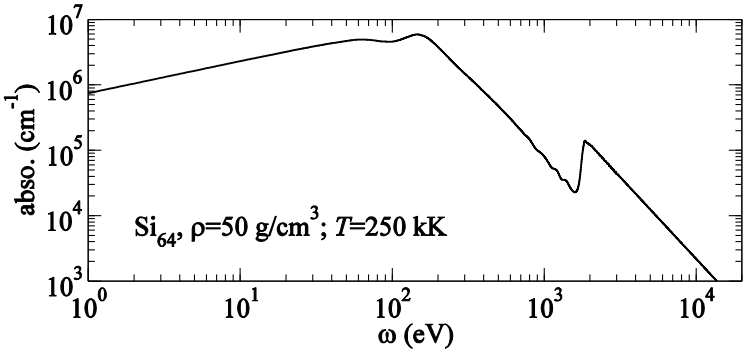


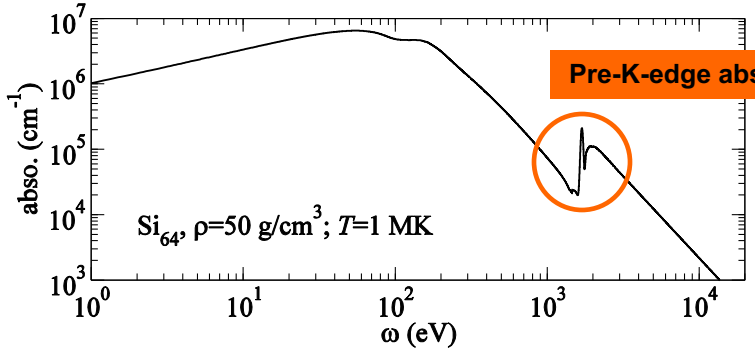


A strong pre-K-edge absorption has been found at high temperatures

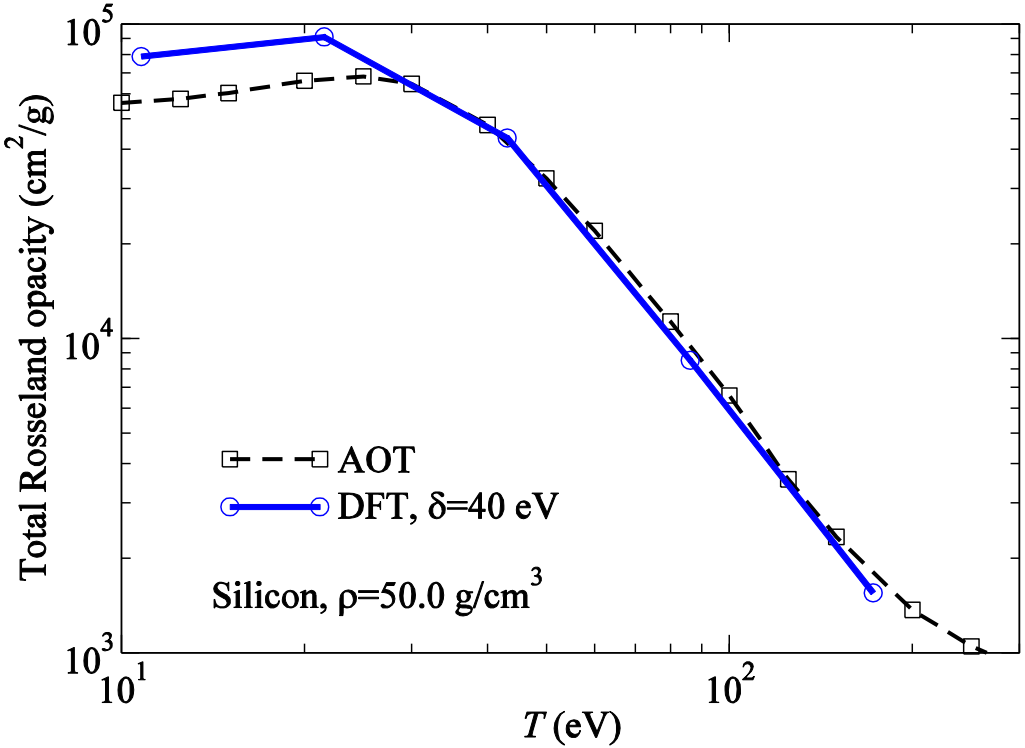








Comparison between the AOT and DFT data for total opacity, silicon ρ=50.0 g/cm^3 K shows discrepancy at T<30 eV.



- AOT data are not accurate at low temperatures relevant to the ICF implosion conditions
- The same trend was previously observed in AOT data for other materials D_2 and CH (Hu, et al. PRE 90, 033111 (2014); Hu, et al. PRB 96, 144203 (2017))

Conclusions

Systematic first-principles calculations of absorption coefficients and opacities of silicon have been performed in a wide range of material densities and temperatures

- Our density-functional theory (DFT)-based calculations naturally account for quantum effects, continuum lowering, and Fermi-surface rising in hot dense silicon plasmas

- A set of accurate all-electron pseudo-potentials to calculate x-ray absorption near edge structure (XANES) spectra has been constructed for first time

- Absorption coefficients and opacities of silicon (densities between 0.1 and 500 g/cm^3 and T between 0.5 and 1000 eV) have been calculated