INL/MIS-23-73708-Revision-0

Anisotropy in Thermal Conductivity of Ga2O3 under Irradiation: Ab-initio Investigations

August 2023

Cheng Sun, Arjun Sathyan Kulathuvayal

Changing the World's Energy Future

Idaho National Laboratory

INL is a U.S. Department of Energy National Laboratory operated by Battelle Energy Alliance, LLC

INL/MIS-23-73708-Revision-0

Anisotropy in Thermal Conductivity of Ga2O3 under Irradiation: Ab-initio Investigations

Cheng Sun, Arjun Sathyan Kulathuvayal

August 2023

Idaho National Laboratory
Idaho Falls, Idaho 83415

http://www.inl.gov

Prepared for the
U.S. Department of Energy
Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517

Anisotropy in Thermal Conductivity of β-Ga_2O_3 under Irradiation: Ab-initio Investigations

Arjun S Kulathuvayal | Utah State University | Mentor : Dr. Cheng Sun , U210 – Advanced characterization and post-irradiation examination division – Material Fuel Complex.

Background

Ga_2O_3 – a wide-bandgap semiconducting material with a high breakdown electric field and thermal stability. Ga_2O_3's crystalline forms are known for promising performance in power electronics, optoelectronics, and high-temperature applications. Particularly, the β-Ga_2O_3 is well established for its potential application in neutron and gamma radiation detection hence can be utilized for sensor and detector applications[1].

β-Ga_2O_3 exhibits anisotropic thermal conductivity, meaning that the magnitude of its thermal conductivity is dependent on the crystallographic direction[2].

Experimental Observations

When Ga_2O_3 is subjected to neutron irradiation, a notable phenomenon occurs where Helium bubbles start to form within the crystalline matrix. This presence of Helium in Ga_2O_3 has been found to have an impact on the material's anisotropic thermal conductivity.

Objectives of Ab-initio DFT Studies

- To identify the most probable Helium occupation sites in β-Ga_2O_3 using Density Functional Theory (DFT).
- To investigate the anisotropic variation in thermal conductivity when Helium occupies in the crystalline matrix of β-Ga_2O_3.

Theoretical Aspects – Phonon Dispersion

- Phonons are **quantized vibrational modes** of the crystal lattice. Dispersion relation for 2D lattice then,

$$M_1\frac{d^2u_s}{dt^2} = C(v_s + v_{s-1} - 2u_s)$$
$$M_2\frac{d^2u_s}{dt^2} = C(u_{s+1} + u_s - 2v_s)$$

For a simple diatomic basis, equation of motion of atoms.

$$\omega^2 = -\frac{C}{M}\left[e^{ika} + e^{-ka} - 2\right]$$

Phonon dispersion relation in general

$$u_1 = ue^{ika}e^{-i\omega t}$$
$$v_2 = ve^{ika}e^{-i\omega t}$$
$$\rightarrow \begin{vmatrix} 2c - M_1\omega^2 & -C\left[1 + e^{ika}\right] \\ -C\left[1 + e^{ika}\right] & 2C - M_2\omega^2 \end{vmatrix} = 0$$

Solution as a traveling wave with amplitude u and v.

Solving this equation for limiting cases ($ka \ll 1$) gives optical and acoustical phonon dispersion spectra.

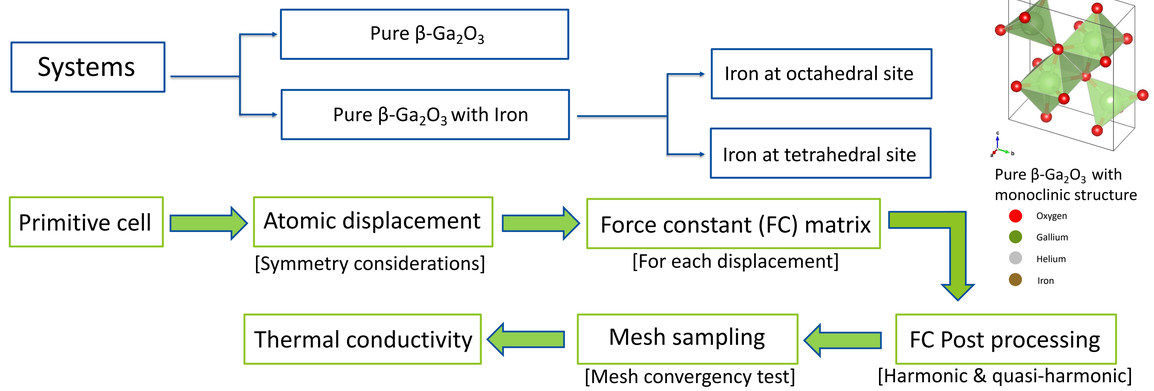
k : wave vector in crystal lattice
a : distance between two atoms
C : lattice force constant ; and M : Reduced mass

- Lattice thermal conductivity studied using **phonon-phonon interaction** in β-Ga_2O_3.
- Phonon interactions were simulated at **harmonic and quasi-harmonic levels**.
- **Finite Displacement Method** were used for calculating the lattice force constant.
- Lattice thermal conductivity computed by **relaxation time approximation** and direct-solution of **phonon Boltzmann equation** and the **Wigner transport equation**.

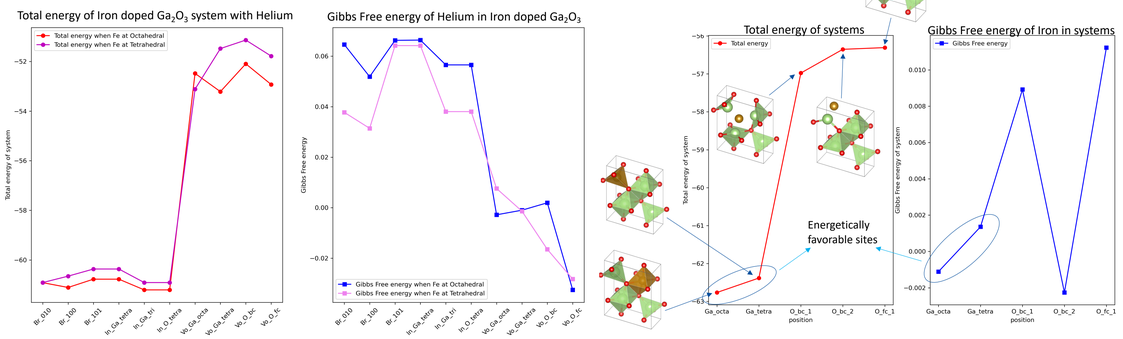
[1] Anisotropic thermal conductivity in single crystal β-gallium oxide; Guo, Zhi and Verma, Amit, Appl. Phys. Lett.; AIP Publishing; 10.1063/1.4916078.

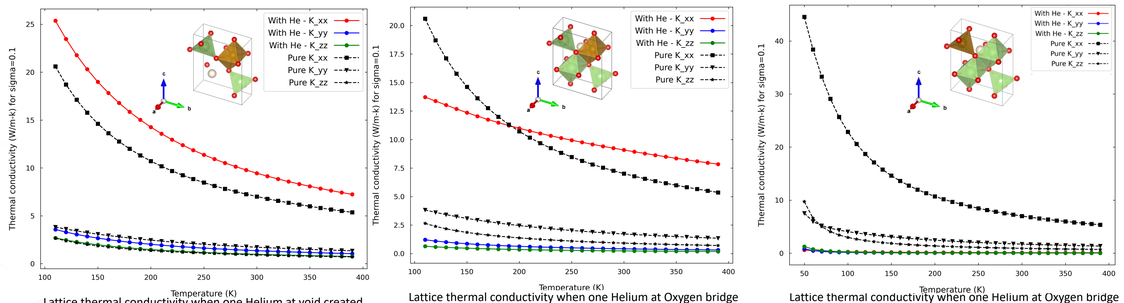
[2] Three-dimensional anisotropic thermal conductivity tensor of single crystalline β-Ga2O3; Jiang, Puqing and Qian, Xin and Li; Appl. Phys. Lett.; AIP Publishing; 10.1063/1.5054573.

Workflow



Results & Conclusions





Lattice thermal conductivity when one Helium at void created by a Gallium atom in Iron doped Ga_2O_3 (Octahedral site)

Lattice thermal conductivity when one Helium at Oxygen bridge sites in [100] direction in Iron doped Ga_2O_3 (Octahedral site)

Lattice thermal conductivity when one Helium at Oxygen bridge sites in [100] direction in Iron doped Ga_2O_3 (Tetrahedral site)

- Anisotropy in thermal conductivity (TC) has observed in β-Ga_2O_3 when Helium occupies in interstitial and lattice spaces.
- Since Iron is present in the experimental sample, its contribution to the TC has been studied and revealing that iron prefers stable occupancy in octahedral voids rather than tetrahedral voids created by Gallium atoms.
- The anisotropy in TC becomes evident when Iron occupies octahedral voids, while it disappears when iron is situated in tetrahedral sites.