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sites in [100] direction in Iron doped Ga_2O_3 (Tetrahedral site)

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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 -Ga₂O₃ 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 -Ga₂O₃ using Density Functional Theory (DFT).
- To investigate the anisotropic variation in thermal conductivity when Helium occupies in the crystalline matrix of β -Ga₂O₃.

Theoretical Aspects - Phonon Dispersion

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

$$M_{1} \frac{d^{2}u_{s}}{dt^{2}} = C(v_{s} + v_{s-1} - 2u_{s})$$

$$M_{2} \frac{d^{2}u_{s}}{dt^{2}} = C(u_{s+1} + u_{s} - 2v_{s})$$

For a simple diatomic basis, equation of

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

Phonon dispersion relation in general

- $\begin{vmatrix} u_1 = ue^{ika}e^{-i\omega t} \\ v_2 = ve^{ika}e^{-i\omega t} \end{vmatrix} \rightarrow \begin{vmatrix} 2c M_1\omega^2 & -C[1 + e^{ika}] \\ -C[1 + e^{ika}] & 2C M_2\omega^2 \end{vmatrix} = 0$
- 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 -Ga₂O₃.
- 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 Pure β -Ga₂O₃ Systems Iron at octahedral site Pure β-Ga₂O₃ with Iron Iron at tetrahedral site Pure β-Ga₂O₃ with monoclinic structure Atomic displacement Primitive cell Force constant (FC) matrix [For each displacement] [Symmetry considerations] Mesh sampling Thermal conductivity FC Post processing [Harmonic & quasi-harmonic] [Mesh convergency test] Results & Conclusions Total energy of Iron doped Ga₂O₃ system with Helium Gibbs Free energy of Helium in Iron doped Ga₂O₃ Gibbs Free energy of Iron in system Total energy when Fe at Octahedral Energetically O_bc_2 Pure K_yy ----Pure K_yy --₹--

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• Anisotropy in thermal conductivity (TC) has observed in β -Ga₂O₃ when Helium occupies in interstitial and lattice spaces.

by a Gallium atom in Iron doped Ga₂O₃ (Octahedral site)

• 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.

sites in [100] direction in Iron doped Ga₂O₃ (Octahedral site)

 The anisotropy in TC becomes evident when Iron occupies octahedral voids, while it disappears when iron is situated in tetrahedral sites.

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