POINT DEFECT PHONON SCATTERING: FUNDAMENTALS AND IMPORTANCE

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INTRODUCTION

The present work deals with understanding the effect of point-defect phonon scattering on the thermal transport properties of semiconductors. Phonons are the main heat carriers in semiconductors and insulators [1]. Various point defects like vacancies, interstitials, and impurities are inherently present in such materials. These defects introduce mass and interatomic force constant (IFC) perturbations that cause a hindrance to the flow of phonons and determine the phonon scattering characteristics of a particular point defect. The higher the scattering caused by a point defect, the larger the induced reduction in the lattice thermal conductivity (κ_ℓ). Experimentally, it is challenging to retrieve the phonon scattering rates of individual defects. However, recent advances on the theoretical and computational fronts have facilitated understanding and dealing with such a problem. In this regard, the T-matrix atomistic Green's functions (AGF) approach combined with inputs from density functional theory (DFT) facilitates the treatment of the perturbation to all orders and parameter-free calculation of scattering rates [2].

AGF have been successfully applied for studying defects like vacancies, extrinsic dopants, nanoparticles, and dislocations [3]. Here we explain the drastic reduction in κ_ℓ of cubic SiC (3C-SiC) in the case of boron doping based on the enhanced phonon scattering it causes [4]. The underlying factors in the AGF that result in such enhanced scattering are studied in detail. It is established that the smaller the asymmetry in the atomic relaxation around the defect, the higher the IFC perturbation, and the higher the phonon scattering [5].

THEORETICAL FOUNDATION

The scattering rate due to point-defect phonon interactions can be calculated as:

$$\frac{1}{\tau_{i\mathbf{q}}} = \pi n_{\text{def}} V_{\text{uc}} \frac{1}{\omega_{i\mathbf{q}}} \sum_{i'\mathbf{q}'} \left| \left\langle i'\mathbf{q}' \left| \mathbf{T} \right| i\mathbf{q} \right\rangle \right|^2 \delta \left(\omega^2_{i'\mathbf{q}'} - \omega^2_{i\mathbf{q}} \right), \tag{1}$$

where where n_{def} is the volumetric concentration of the point defects, V_{uc} the volume of the unit cell, and ω the angular frequency of phonons. The sum is taken over all the possible outgoing phonons of wave vector $\mathbf{q}'$ and branch index i' for an incoming phonon $i\mathbf{q}$ scattered from a point defect. The $\mathbf{T}$ matrix is defined as, $\mathbf{T} = (\mathbf{I} - \mathbf{V}\mathbf{g}^+)^{-1}\mathbf{V}$, where $\mathbf{g}^+$ is the retarded phonon Green's function of the unperturbed host lattice. $\mathbf{V}$ is the total perturbation matrix calculated as, $\mathbf{V} = \mathbf{V}_M + \mathbf{V}_K$. Here, $\mathbf{V}_M$ and $\mathbf{V}_K$ are the mass and IFC perturbation matrices, respectively.

RESULTS AND DISCUSSION

In 3C-SiC, we observed that nitrogen doping in polycrystalline samples reduces κ_ℓ only slightly as compared to the undoped ones, in excellent agreement with experiments [4]. However, addition

of boron to the system in amounts much smaller than nitrogen leads to a huge reduction in κ_ℓ. This reduction is caused by the enhanced scattering rates of the neutral B defect as compared to N, see Fig. 1, which shows the scattering rates of B- and N-doped 3C-SiC and diamond in different charge states. Boron shows resonant phonon scattering at ≈ 33 rad/ps and the scattering rates are two orders of magnitude larger than the N ones. This was found to be directly related to the fact that the B atom relaxes slightly asymmetrically, away from one of the Si atoms in the nearest neighbor Si atom tetrahedron around it, as shown in Fig. 2. This resulted in a large IFC perturbation which is essential for enhanced scattering. However, for B- and N-doping in diamond, it is observed that the N atom relaxes highly asymmetrically and causes an IFC perturbation smaller than B which relaxes slightly asymmetrically. Despite producing equally large IFC perturbations no enhanced scattering is observed in diamond because of its low phonon density of states.

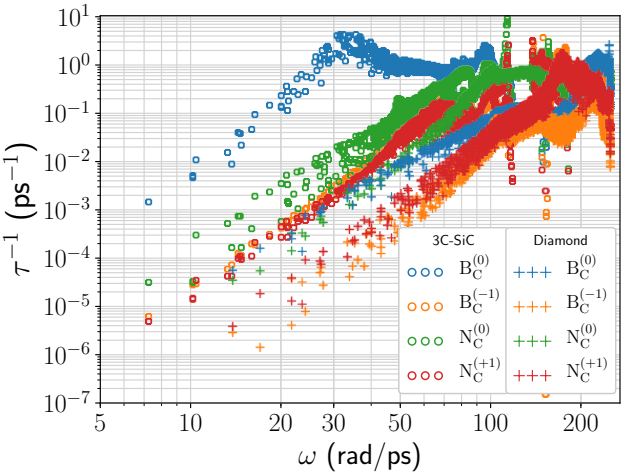

Figure 1: Scattering rates of different dopants in 3C-SiC and diamond [4]-Reproduced by permission of The Royal Society of Chemistry.

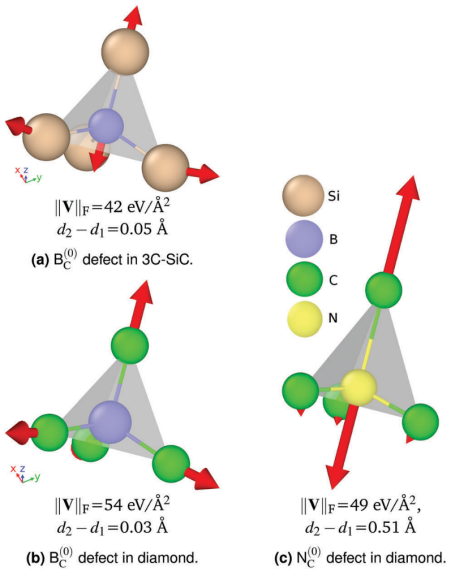

Figure 2: Asymmetrical relaxation of different defects in 3C-SiC and diamond [5]-Reproduced by permission of The Royal Society of Chemistry.

CONCLUSION

We have shown how certain point defects scatter phonons more strongly than others and can greatly reduce κ_ℓ. Asymmetrical relaxation of the defect in the system is an important ingredient to resonant scattering. In future, we plan to perform a high-throughput study of all the possible points defects in many different semiconductor materials with an aim of identifying other such super-scatterers.

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