# Microscopic origin of heat properties in ScN epitaxial film revealed by inelastic X-ray scattering spectroscopy

A challenge of next-generation devices (e.g., solar cells, thermoelectric devices, power devices) is to reduce the amount of energy wasted. In power devices, for instance, the applied high electric field easily causes overheating, resulting in operating failures. In thermoelectric devices and solar cells, lower energy loss is required for higher efficiency. In particular, in the coming years, these devices will be required to be compact while maintain their performance, and hence detailed knowledge of their heat properties and thermal management will be important for designing and fabricating these devices.

Phonons are the particles that carry heat in semiconductors and insulators. These particles have infinite lifetime in the harmonic approximation. However, once a particle merges with another particle or decays into two particles (as shown in Fig. 1(a)), the harmonic approximation is no longer satisfied, and the phonon has a finite lifetime. Thermal conductivity is directly correlated to the phonon lifetime; in other words, a (in)finite phonon lifetime corresponds to (in)finite thermal conductivity. On the one hand, in power device applications, good thermal conductivity (i.e., a long phonon lifetime) is desired. On the other hand, in thermoelectric applications, poor thermal conductivity (a short phonon lifetime) leads to a good thermoelectric figure of merit.

The phonon lifetime is a function of the phonon mode and momentum. The recent development of density functional theory (DFT) calculations now provides both the microscopic phonon lifetime, including the phonon-mode and momentum dependences, and the macroscopic thermal conductivity. Experimentally, the phonon lifetime with momentum dependence at a specific phonon mode has been estimated by observation of the phonon linewidth in inelastic neutron and X-ray scattering profiles [1,2]. Compared with neutron scattering, the attenuation length of X-rays is generally much smaller (<~1 mm), and tunable by changing the incident angle ($\alpha$, Fig. 1(b)), which enables measurements of the phonon properties of films. In the previous literature, however, phonon lifetimes of bulk materials were estimated, and there have been no reports of the momentum-dependent phonon lifetime in epitaxial films.

We have observed the phonon lifetime of ScN, a promising thermoelectric material among the nitride semiconductors, using high-resolution inelastic X-ray spectroscopy (IXS) at SPring-8 **BL35XU** [3]. This material has a rocksalt structure, in which no phonon modes should be observed in the first-order Raman scattering, and therefore detailed phonon information remains unknown. In this study, we used an epitaxially grown ScN (110) film (thickness of 40 μm) on a sapphire substrate (m-plane). After optimizing the incident angle with respect to the surface of the film ($\alpha = 1.1$–$1.7°$ was used in the measurements), we obtained information only from the ScN film without artifacts from the substrate (as shown in Fig. 2, these spectra correspond to the phonon dispersion relation along the [$q$00] (right) and [$qq$0] (left) directions). Note that the obtained spectra in Fig. 2 are well reproduced by the DFT calculations.

To discuss the phonon lifetime in ScN, we estimated the profile linewidth of the longitudinal optical phonon mode along the [$q$00] direction (black dots in Fig. 3(a). The corresponding phonon dispersion is indicated by red arrows in Fig. 2). The phonon lifetime is short near the $\Gamma$ point (around 0.2 ps) but long around the X point (more than ~3 ps). This indicates that phonon scattering (Fig. 1(a)) frequently occurs at $\Gamma$ but rarely happens at X. Similar to the phonon dispersion in Fig. 2, the phonon lifetime is also reproduced by the DFT calculations (red line in Fig. 3(a)). Owing to this agreement, we can obtain the detailed phonon scattering process as shown in Figs. 3(b-d). Similar agreement between observation and calculations is obtained in the transverse phonon mode along the [$q$00] direction (the corresponding phonon dispersion is indicated by black arrows in Fig. 2).

The agreement between the experimental observation and DFT calculations of the phonon lifetime enables us to discuss the macroscopic thermal conductivity; the DFT calculation indicates that the thermal conductivity is 43 W/m/K at $T = 300$ K, which well reproduces the observed thermal conductivity, 36.4 W/m/K. This good agreement indicates that

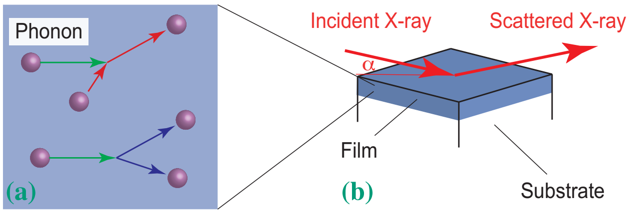

Fig. 1. **(a)** Phonon-phonon scattering mechanisms that reduce the thermal conductivity. **(b)** Schematic of experimental setup; thin film on substrate and path of X-ray used in the experiment.

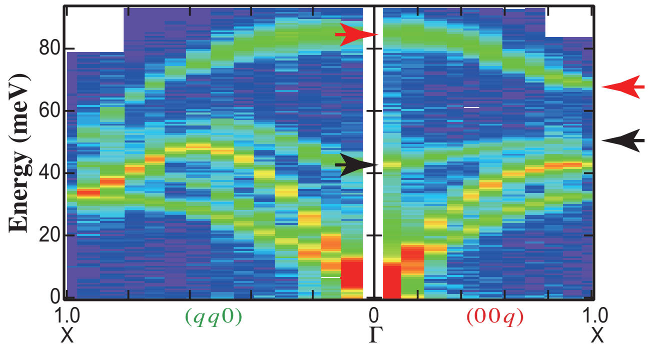

Fig. 2. Phonon dispersion curves in ScN film observed by inelastic X-ray scattering spectroscopy.

the origin of macroscopic thermal conductivity can be explained by the macroscopic phonon scattering mechanism as shown in Fig. 1(a). In the previous reports, this material has relatively low thermal conductivities of 10–20 W/m/K. However, our study reveals that the material has potentially higher thermal conductivity from the viewpoints of both observation and calculations. Through the comparison of the carrier and thermal conductivities in the literature, we found that impurities and defects may have reduced the thermal conductivity in the previous reports.

The present work demonstrates that the microscopic mechanism of thermal properties in a thin film, which is similar to an actual device environment, can be clarified by comparing the IXS profiles with the results of thermal conductivity measurements and calculated data. The achievements of this study are expected to be used to evaluate the thermal properties of leading-edge energy-efficient/energy creation devices such as thermoelectric devices that convert heat into electricity, highly efficient power semiconductor devices with low energy loss, and next-generation solar cells.

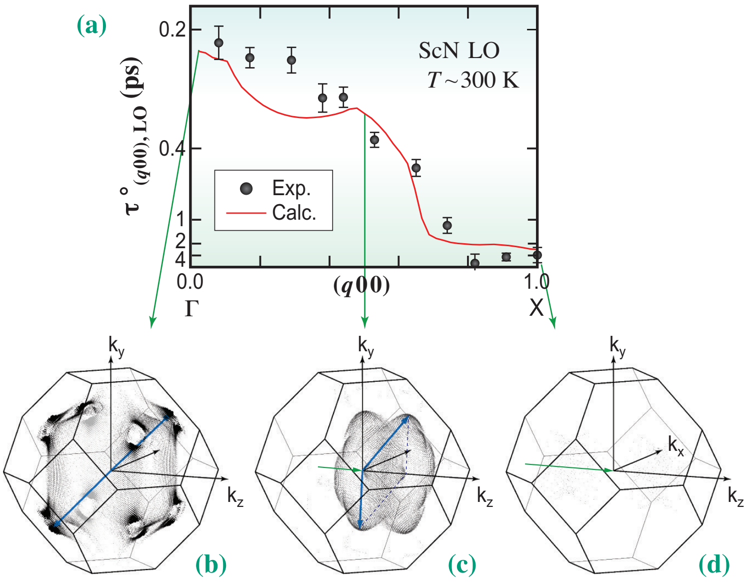

Fig. 3. **(a)** Experimentally obtained phonon lifetime (black solid circles) and lifetime in the DFT calculations (red line). **(b)-(d)** Plot of the calculated phonon-phonon scattering mechanism. The blue arrows in (b) and (c) show one pair of decay phonons from the original ($q$00) phonon (green arrow). Near the X point, phonon scattering (black dots) rarely occurs, as shown in (d). Blue and green arrows in (b)-(d) correspond to the phonon scattering mechanism depicted in Fig. 1(a) with the same colors.

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**References**

[1] O. Delaire *et al.*: Nat. Phys. **10** (2011) 614.
[2] D.J. Voneshen *et al.*: Nat. Mater. **12** (2013) 1028.
[3] *H. Uchiyama, Y. Oshima, R. Patterson, S. Iwamoto, J. Shiomi and K. Shimamura: Phys. Rev. Lett.* ***120*** *(2018) 235901.*