Estimation of phonon lifetime in epitaxial films

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Phonon measurements in SPring-8

IXS BLs for phonon measurements

BL35XU & BL43LXU

Both are design and built by A. Baron

SPring-8

BL35XU

Public facility:

~4000 hours/year for Users

Heat and Phonons

Heat and phonon

In recent device applications,

Controlling heat properties are now important

as controlling electronic properties.

1. Energy saving/harvesting

   Heat is unused (wasted) energy;

   it should be used more efficiently..

   e.g. thermoelectric, photovoltaic devices

2. Preventing overheating

   due to miniaturization and power application

   heat can damage the specification of devices…

In order to control heat properties,

⇨ Phonon properties

should be investigated

Heat and phonon

For example,
(lattice) thermal conductivity is given by

$$\kappa_{lat} = \frac{1}{3NV_0} \sum_{\mathbf{q},j} \underbrace{C_{\mathbf{q},j}}_{\text{Specific heat}} \underbrace{\mathbf{v}_{\mathbf{q},j}}_{\text{Phonon velocity}} \otimes \mathbf{v}_{\mathbf{q},j} \underbrace{\tau^{\circ}_{\mathbf{q},j}}_{\text{Phonon lifetime}}$$

Especially phonon lifetime can not be produced by the harmonic approximation.

⇨ Consideration beyond harmonic approximation is required

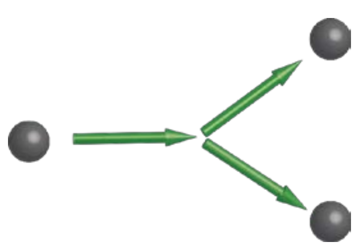
Phonon lifetime $\tau^{\circ}_{\mathbf{q},j}$ can be calculated and measured

In calculation, (for example, phonon-phonon interaction),

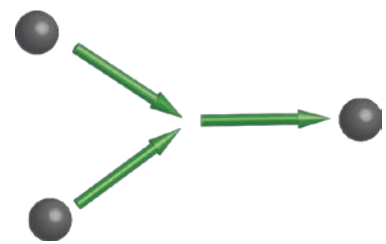
Phonon lifetime can be given as a imaginary part of phonon self-energy ($\Sigma^{\circ}_{\mathbf{q},j}$)

$$\tau^{\circ}_{\mathbf{q},j} = \hbar / 2\mathrm{Im}\Sigma^{\circ}_{\mathbf{q},j}$$

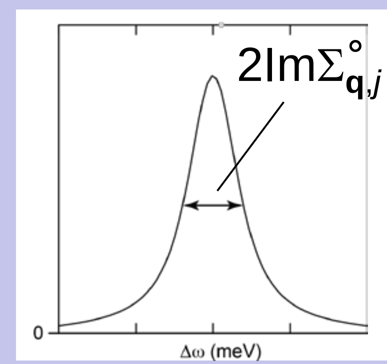
In the lowest order, $\tau^{\circ}_{\mathbf{q},j}$ is given as three–phonon interaction



Decay Process



Merging Process



In measurements,

Spectral line width corresponds to $2\mathrm{Im}\Sigma^{\circ}_{\mathbf{q},j}$:

estimated by IXS, INS, and Raman

⇨ Macroscopic thermal properties can be compared to microscopic phonon measurements

IXS for epitaxial films

IXS for epitaxial films

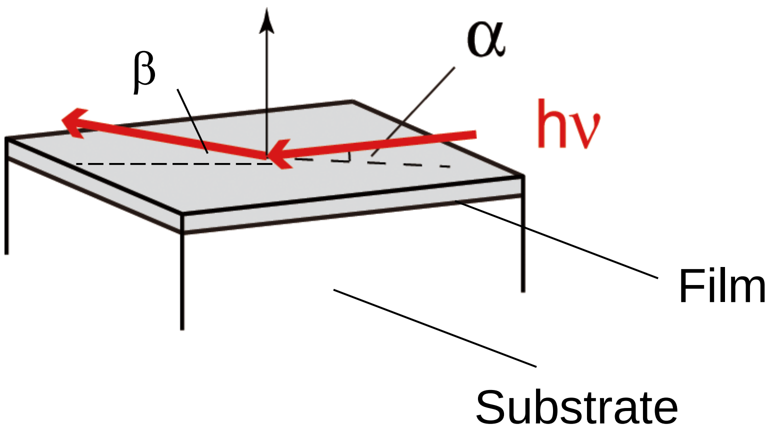
Phonon lifetime can be measured spectral width of

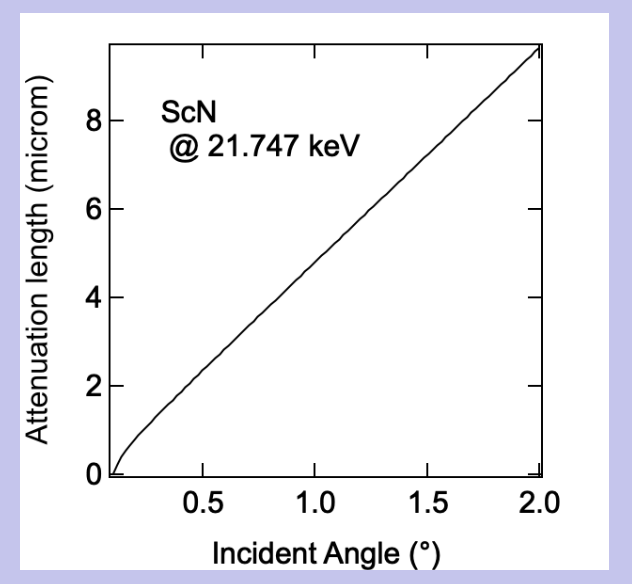
- ✓ INS (long probing depth)
- ✓ Raman (only at Γ)

Not suitable for epitaxial film with momentum dependence
(cf. definition of thermal conductivity)

IXS

Proper incident beam angle (α) only penetrates only film.



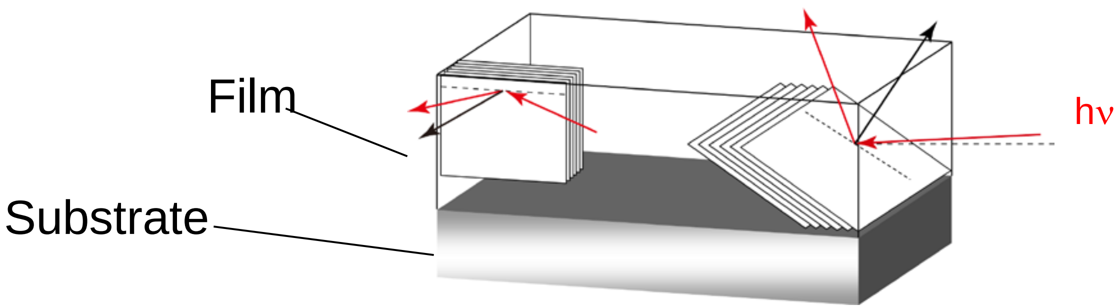


IXS for epitaxial films

Two configurations for achieving low incident angle (cf. GIXRD)

In plane
Symmetry ($\alpha \sim \beta$)

Out of plane (Brennan)
Asymmetry ($\alpha << \beta$)



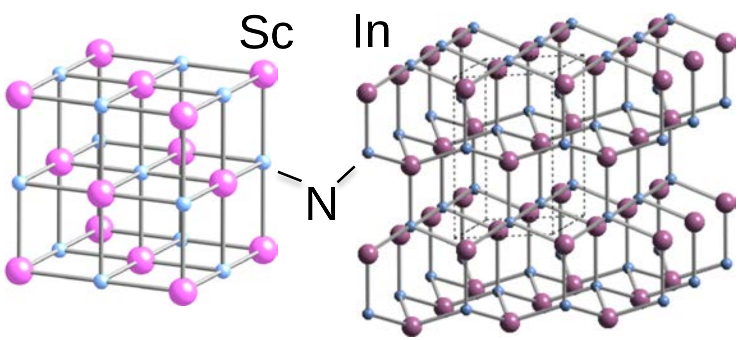
	In plane	Brennan
Bragg	In the plane	Not in the plane
Setup	Easy	Difficult (needs math)
Precision	Small kz component	Accurate
Changing incident angle	Difficult	Easy

S. Brennan, Physica B(283) 125 (2000)

IXS for epitaxial films

Nitrides

For application of
- power device,
- light emitting device,
- photovoltaic device,
- etc.



ScN & InN：mainly available as thin films (not bulk materials)

ScN; thermoelectric device

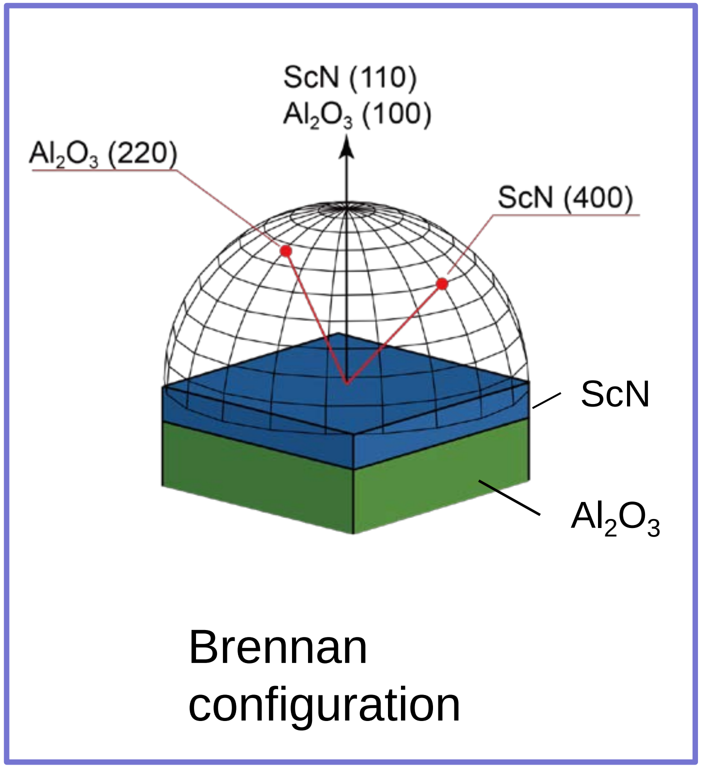
ZT～0.3 (at 800 K)*

InN; phonon gap, phonon bottleneck

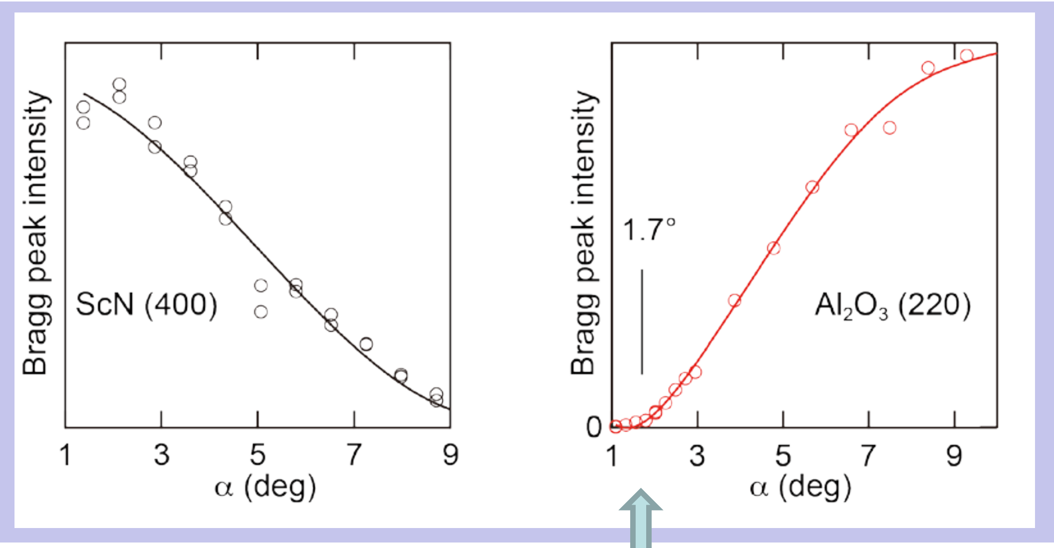
S. Kerdsongpanya, APL(99) 232113

Example 1: ScN (40 μm)/ Al_2O_3

Incident beam angle dependence for Bragg peaks



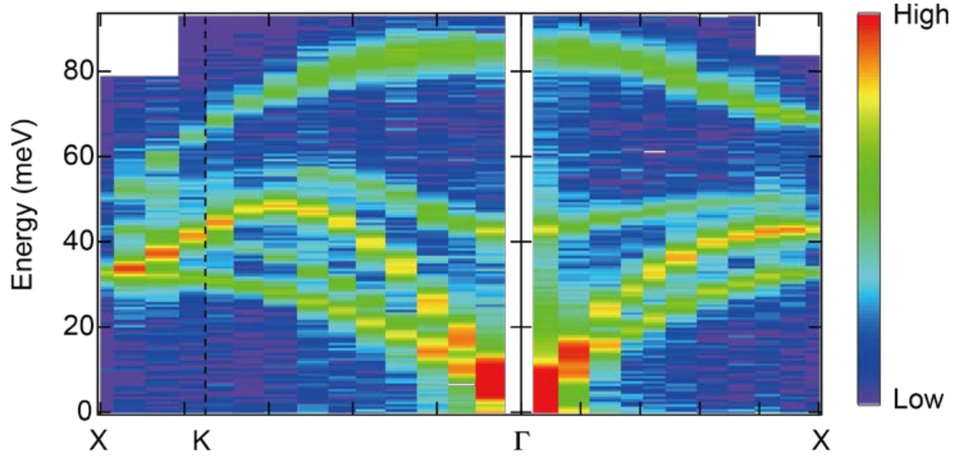
Bragg peak intensity measurements



At α<1.7°
Substrate contribution is negligible

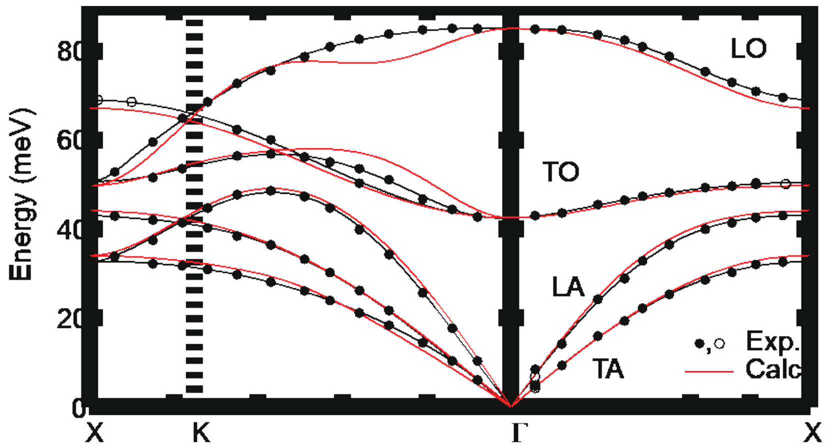
Example 1: ScN (40 μm)/ Al_2O_3

IXS spectra (logarithmic scale)



ScN RT
$h\nu$=21.747 keV
(ΔE=1.4~1.7 meV)
α=1.1~1.7°

Comparison with DFT



Good agreement

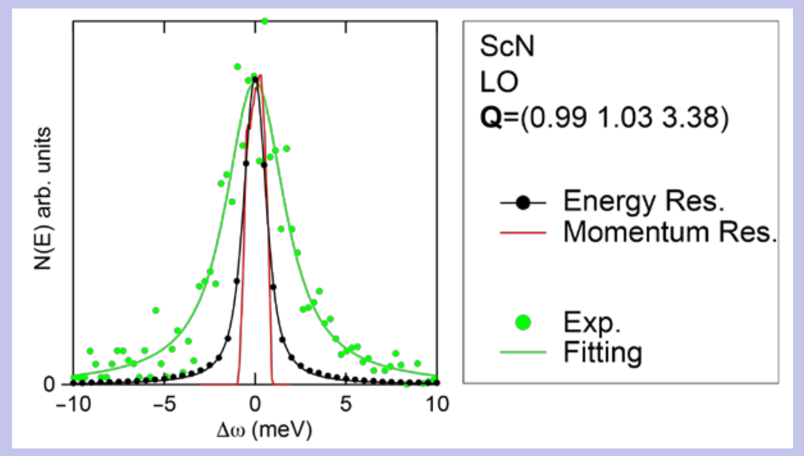
No contribution from substrate

HU, PRL(120) 235901

Estimation of phonon lifetime

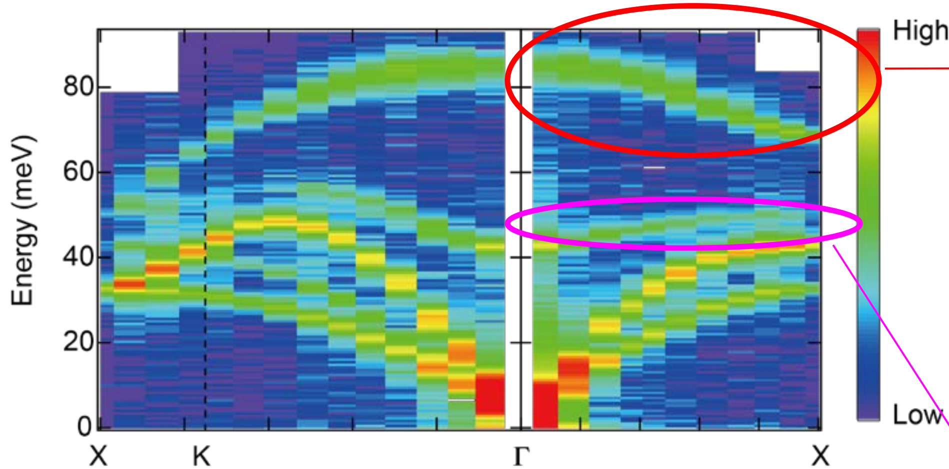
How to determine anharmonic contribution accurately?

An IXS profile (linewidth) includes conributions from
- Energy resolution
- Momentum resolution
- **Anharmonic contribution** ⟸ should be extracted

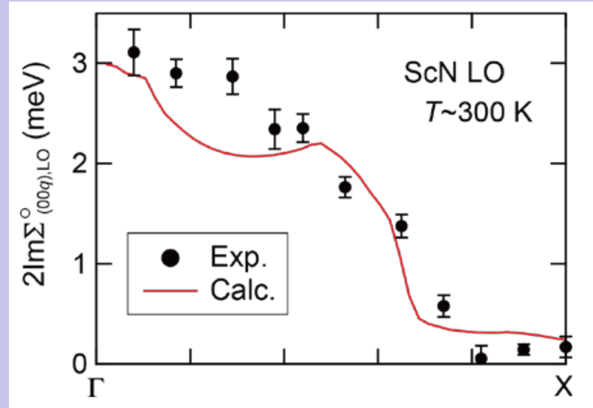


Estimation of phonon lifetime

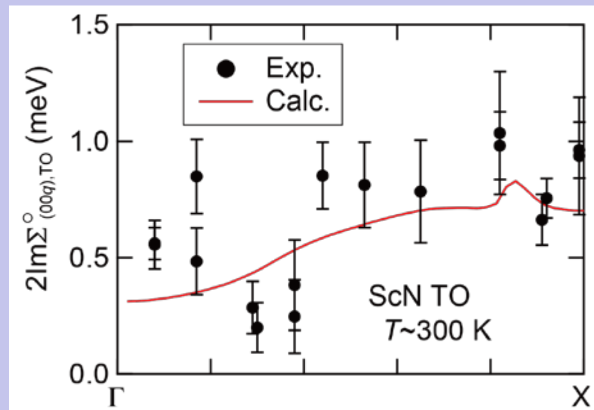
IXS spectra



Lifetime of LO along Γ-X



Lifetime of TO along Γ-X



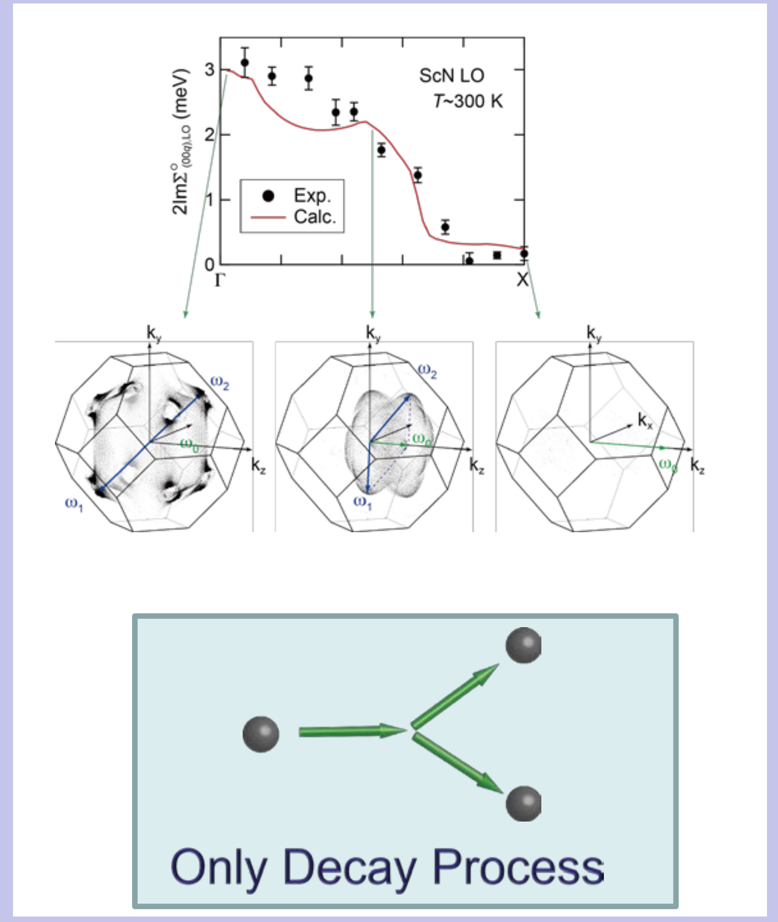
Phonon lifetime is also reproduced by calculation **(three-phonon interaction)**

Estimation of phonon lifetime

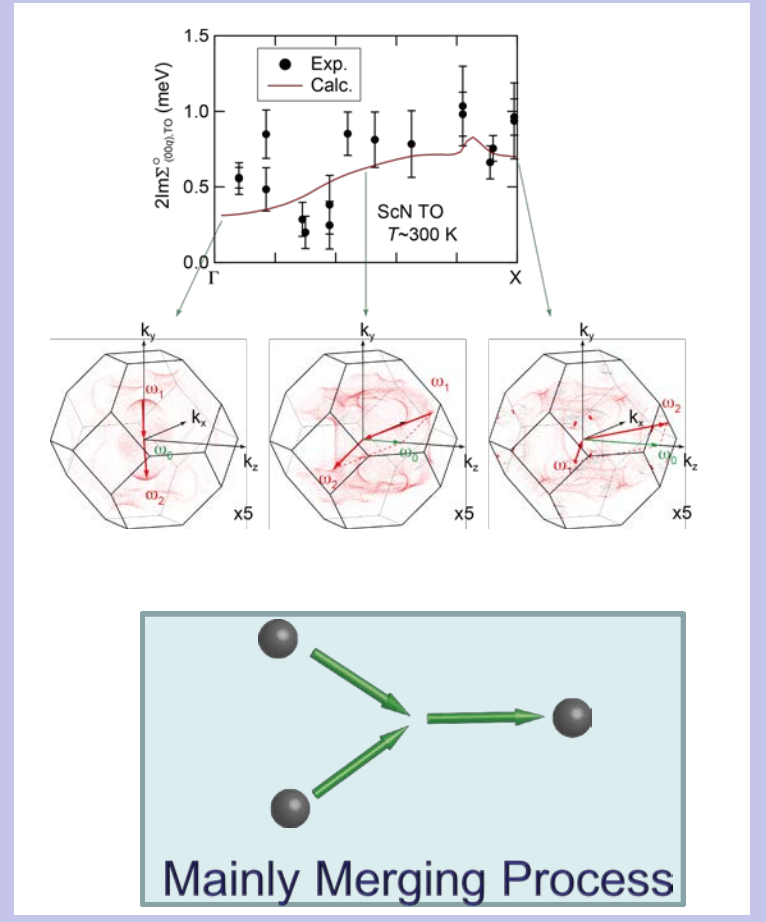
Good agreement (between Exp. and Calc.) of phonon lifetime in ScN

Detailed scattering process can be displayed

Lifetime of LO along Γ-X



Lifetime of TO along Γ-X



Estimation of phonon lifetime

Furthermore, calculation indicates the macroscopic thermal conductivity

$$\kappa_{lat} = \frac{1}{3NV_0} \sum_{\mathbf{q},j} C_{\mathbf{q},j} \mathbf{v}_{\mathbf{q},j} \otimes \mathbf{v}_{\mathbf{q},j} \tau^{\circ}_{\mathbf{q},j}$$

⇨ 43 W/m/K (at 300 K). Calc.
36.4 W/m/K (at RT). Measured

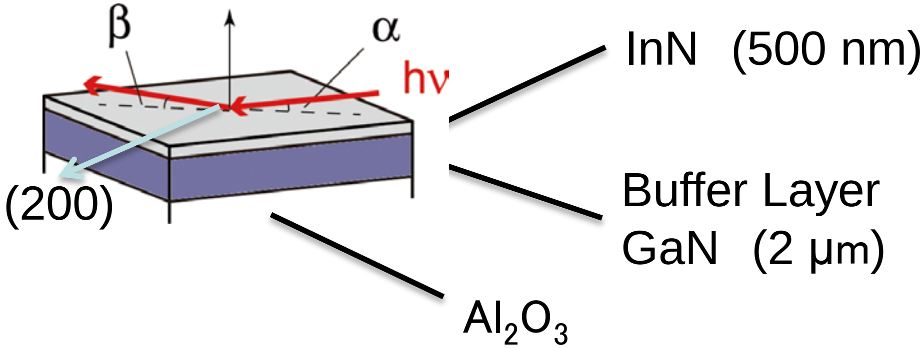
Thermal conductivity also agrees

All heat properties can be reproduced by microscopic phonon properties

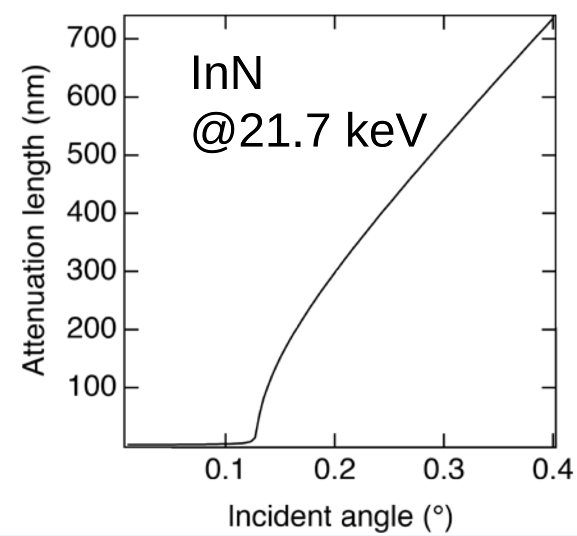
Example 2: InN (500 nm)/ Al_2O_3

Example 2: InN (500 nm)/Al_2O_3

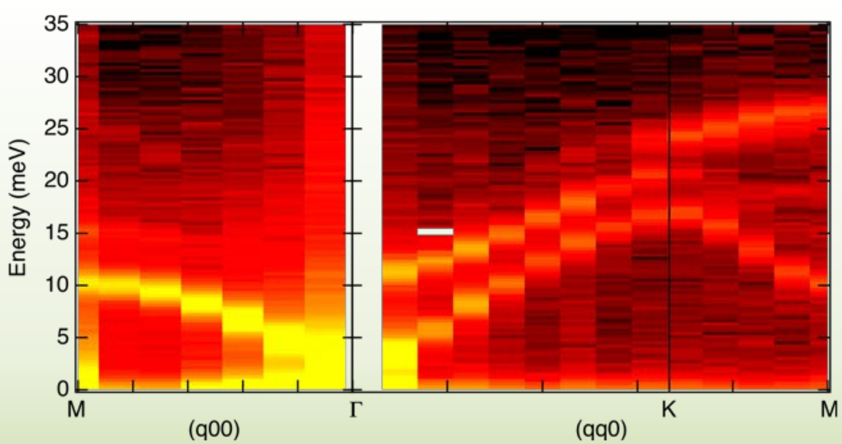
For further thinner：InN (500 nm)



In plane geometry



Transverse phonon



α =0.13°

Uchiyama, Araki in preparation

Summary

◆ In order to control thermal properties?

⇒ Investigating phonon properties provides microscopic mechanism of thermal properties

◆ IXS is a unique method for investigating the phonon properties in epitaxial film

⇒ It reveals

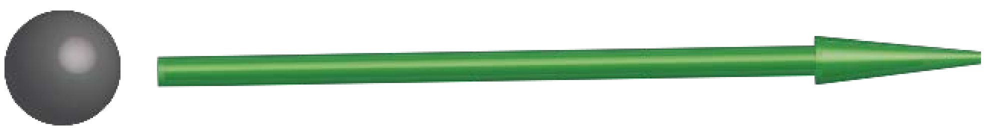
Phonon dispersion and lifetime

using

In-plane and Brennan configurations

Phonon lifetime

in the harmonic approximation

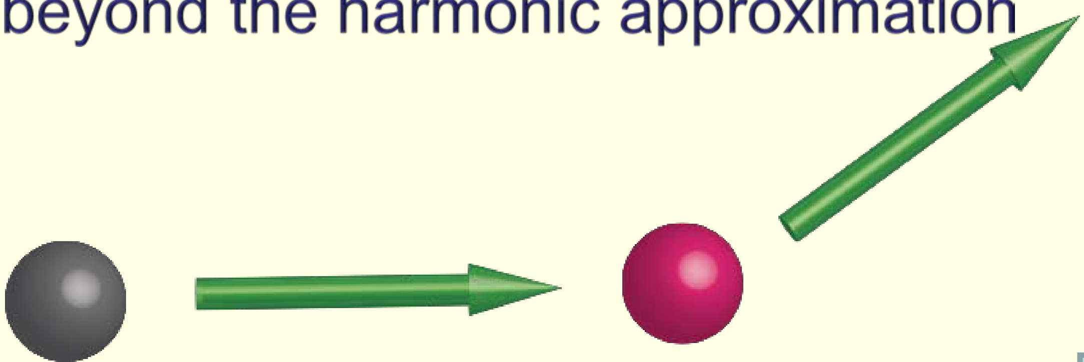


j-th phonon
mode at **q**

Infinite lifetime

⇨ Infinite thermal conductivity

beyond the harmonic approximation



Another phonon,
electron,
boundary…

Finite lifetime

⇨ finite thermal conductivity

Heat and phonon

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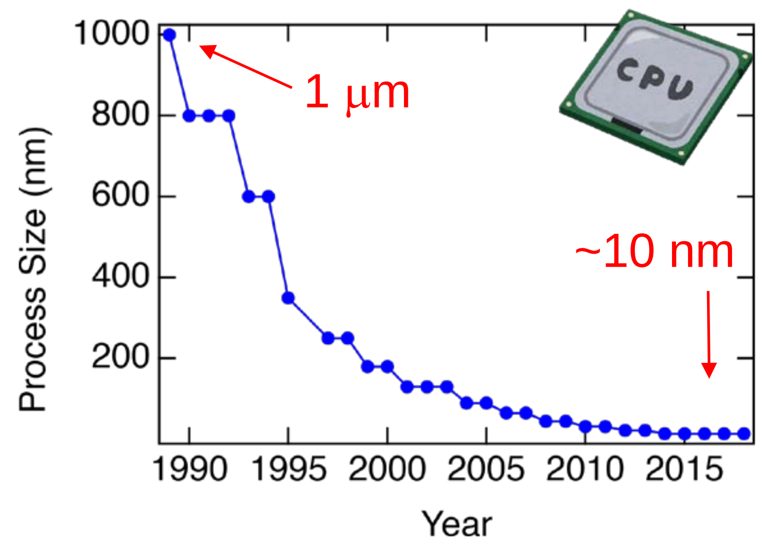

Process size of intel CPU

In order to control heat properties,

⇨ **Phonon properties**

should be investigated