The microscopic phonon properties we have seen are located in small parts of specific paths or planes in the Brillouin zone. Considering that the lattice thermal conductivities are an integrated property over the Brillouin zone, we should investigate the phonon properties taken from the entire Brillouin zone. As such information, in Fig. x are shown phonon densities of states (DOS), cumulative thermal conductivities and their frequency derivatives, weighted DOS with the squares of the group velocity components (vλ,x and vλ,z), and finally, distributions of Γλ．

Firstly, we relate DOS (Fig. x-a) with the cumulative thermal conductivity (Fig. x-b). In α- and γ-Si3N4, the phonons contributing to the lattice thermal conductivities are mainly located below the 1st peaks indicted by an arrow in Fig. x-a, indicating that the main heat carriers are the phonons on the acoustic branches. In contrast, almost a half of the contributions to the zz component of lattice thermal conductivity in β-Si3N4 are derived from the phonons above the 1st DOS peak, indicating that the low frequency optical phonons contribute exceptionally largely to this component.

Secondly, in Figs. x-b and c, the directional differences in the derivatives of the cumulative thermal conductivities in α and β phases are well consistent with the directional differences in the weighted DOS. The relatively larger intensities in the weighted DOS with vλ,z in β-Si3N4 critically causes the large anisotropy in its lattice thermal conductivities.

Γλ in Fig.x-d show similar distributions between the α and β phases, which let the group velocities play a decisive role on the different degrees of the anisotropy of LTC in the two phases. It might be curious that Γλ are similar although group velocities have marked differences. Thus we investigate this similarity further. In Eq.(1) of Γλ, the incorporated term Φ-λλ’λ’’ contains the selection rule for the momentum conservation[Togo], while the selection rule for the energy conservation is expressed by the δ function in Eq. (1). We separately examine the impacts of Φ-λλ’λ’’ on Γλ and the impacts of the selection rules. In Table. xx, the three-phonon interaction strengths are compared as the averages over the frequency ranges between 0 and 15 THz and 0 and 35 THz. The values of the α and β phases are very close to each other, concluding that the three-phonon interaction strengths have similar impacts on Γλ of the α and β phases .

In order to analyze the impacts of the selection rules on Γλ, we employ the joint density of states (JDOS),

(JDOS eq.)

Fig.xx shows the JDOS at three different q points. Although Eq. (1) includes Bose-Einstein functions for the involved phonon modes and JDOS can be weighted with these functions, as in ref. Togo and Scirep, they are omitted for simplicity. With the weights, the absolute values are affected but the weighted JDOS of the α and β phases are still similar. At the low frequency region responsible for the lattice thermal conductivities, among the two terms D(1) and D(2) in (JDOS eq.), dominant is D(2) which corresponds to the half part (ω>= 0) of the auto-correlation function of DOS. The DOS of the α and β phases in Fig. x-a commonly show the frequency gap. The auto-correlation functions, D(2), reflect this DOS feature, dropping suddenly around 0 THz and showing a small shoulder around 7 THz, which corresponds to the width of the gap. Moreover D(2) shows a broad peak around 18 THz, which corresponds to the frequency shift to make the largest correlation between the higher and lower portions of DOS across the gap. Because the gap is basically originated from the differences in vibrations of the planer NSi3 commonly existing in the α and β phases [Kuwabara], the major shapes of D(2), reflecting this DOS feature, are similar in these phases. With the same origin, the JDOS of D(1) are also similar in these phases. With these similar impacts of Φ-λλ’λ’’ and JDOS on Γλ, Γλ in Fig. xx-d are similar in the α and β phases.

As a small difference in Γλ between these phases, Γλ below 5 THz in the β phase are scattered onto two different lines, while those in the αphase are aligned on a smooth line. We investigate the characters of the phonon modes responsible for the behavior of Γλ in the β phase. In Fig.xxx-a, Γλ are classified using colors according to the sums of the squares of the eigenvector components along q; the sum is 1 for perfectly longitudinal waves. However, these sums have no clear contrast to distinguish the two branches in the β phase. Fig.xxx-b shows the same plot as Fig.xxx-a, but with colors according to the sums of the squares of the eigenvector components along the ab plane, which has 1 when the eigenvectors on the ab plane. There is a tendency in the β phase that Γλ are large for the vibrations along the ab plane. Therefore, in the β phase for the phonon modes in the low frequency range below 5 THz, all of which belong to the acoustic phonon branches, the vibration modes along the ab plane are more easily scattered, no matter whether they are longitudinal or transverse. In Fig. xxxx-b-2 a straight line is drawn to divide the phonon modes into two groups. The numbers of the phonon modes in the upper and lower parts are 357 and 126, which are reasonable as the numbers of the vibration modes along the ab plane and out of the ab plane.