The microscopic phonon properties we have seen are located in specific paths or planes in the Brillouin zone. In order to more rigorously inspect the lattice thermal conductivities, we examine phonon properties taken over the Brillouin zone. As such properties, 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, frequency 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, more than 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 to this component exceptionally largely.

Secondly, in Figs. x-b and c, the directional differences in the derivatives of the cumulative thermal conductivities in α and β phases are qualitatively 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.

Fig.x-d shows significantly similar Γλ distributions between the α and β phases, which let the group velocities alone play a decisive role on the different degrees of the anisotropy in the lattice thermal conductivities. Since it might be curious that Γλ are similar although group velocities have marked differences, we investigate this similarity further. Recently Togo et al. Showed that peaks in imaginary part of self energy, Γ(ω), which gives Γλ at ω= ωλ, are mainly brought about by the three phonon selection rules [Togo]. In Eq. (1) of Γλ, the three-phonon interaction strength 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 those of the selection rules, although both of them are not isolated from each other. In Table. xx, Φ-λλ’λ’’ are compared as the averages over the ωλ 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, indicating that the Φ-λλ’λ’’ 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 α phase are aligned on a smooth line. We investigate the characters of the phonon modes responsible for this differnece. 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, for the phonon modes below 5 THz, all of which belong to the acoustic phonon branches, the vibration modes along the ab plane are more easily scattered in the β phase, 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.