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Lattice thermal conductivity in isotope diamond asymmetric superlattices

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1. Introduction

In recent decades, superlattice has been studied in various fields, including electronics [1], optics [2], and phonons [3]. The periodic structure in a superlattice produces unique physical properties, especially in electronic and optoelectronics. Thermal conduction in superlattices has been studied for the purpose of developing a thermal-conductivity controllable material.

Watanabe *et al.* [4] used a chemical-vapor-deposition method to produce $^{12}C/^{13}C$ superlattice diamond thin films and discovered special electrical property, which cannot be explained by individual monolayer characteristics: The excitonic recombination in ^{13}C diamond layer disappears despite that ^{13}C diamond is involved. Such a specific property of $^{12}C/^{13}C$ superlattice may appear in the phonon property like thermal conductivity. It is reported that thermal conductivity is enhanced by nearly 50% by decreasing the isotope impurity (^{13}C) ratio to 0.1% from natural ratio of 1.1%; [5]. Owing to the extremely high thermal conductivity of isotopically pure diamond, it will be possible to control the thermal conductivity of isotope diamond superlattice in a very wide range, because superlattice structure can decrease its thermal conductivity [6]. However, the detail mechanism on the phonon property of diamond superlattice thin film remains unclear. Especially, that for asymmetric isotope diamond superlattices has never been studied.

In this study, we study phonon propagation in isotope superlattices with various lattice period, including asymmetric structures using a lattice dynamics calculation. We further measure the thermal conductivity of diamond superlattices using picosecond ultrasound to confirm the theoretical calculation.

2. Methodology

We measured epitaxial [100] single layer isotope diamond specimens with 99.999% ^{12}C and 99% ^{13}C. We also prepared epitaxial [100] $^{12}C/^{13}C$ superlattice diamond specimens with 30 nm and 100 nm. We use Ti-Sapphire pulse laser with 140 fs duration and 80 MHz repetition rate. The wavelength of the pump and the probe light pulses are 800 nm and 400 nm, respectively. Details of our optics are shown elsewhere [7,8].

For theoretically investigating thermal conduction in superlattices, we perform a theoretical simulation of the lattice thermal conductivity calculation. It starts from phonon kinetic theory:

$$\kappa = \Sigma_\lambda \kappa_\lambda = \Sigma_\lambda C(\omega) v_{\lambda,z}^2 \tau_\lambda, \quad (1)$$

where λ is the phonon mode included the wave number (k_x, k_y, k_z), frequency ω, and polarization j. The total thermal conductivity is obtained from summation of contributions of all phonon modes. The specific heat $C(\omega)$ can be calculated by quantum harmonic oscillator and Planck distribution law. The out-of-plane group velocity $v_{\lambda z}$ can be obtained by lattice dynamics. The phonon mean free time τ_λ is highly ambiguous and we evaluated the normalized thermal conductivity κ' by assuming that phonon mean free time is same at each mode.

$$\kappa' = \Sigma_\lambda C(\omega) v_{\lambda,z}^2 \tau. \quad (2)$$

Such simplification is proposed by Tamura *et al.* [6] In our model, each atom is connected to the 4 first nearest neighbor atoms by a bond-stretching K_1 and bond-bending γ_1 and 12 second nearest neighbor atoms by a bond-stretching K_2 and bond-bending γ_2. The layer numbers of the ^{12}C and ^{13}C are labeled as (n_{12C},n_{13C}), respectively.

3. Experiment

Figure 1 shows the as-measured signal for ^{12}C diamond thin film. The out-of-plane thermal conductivity is evaluated by the background attenuation because it is proportional to the surface temperature according the photo-thermal effect. Detail of the calculation of thermal conductivity is shown elsewhere. [7,8]

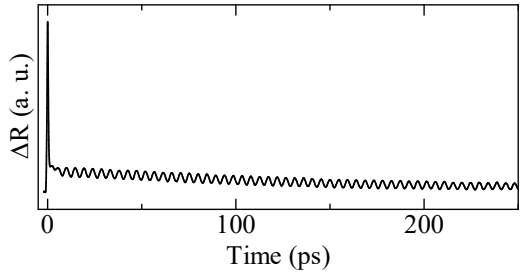

Fig. 1 The typical measured probe light reflectivity change of ^{12}C diamond.

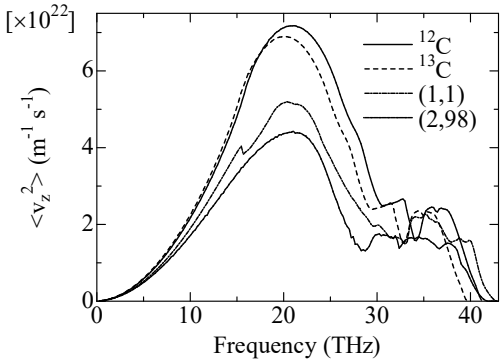

Fig. 2 Phonon DOS weighted by the square out-of-plane velocity.

4. Result & Discussion

From the calculation of lattice thermal conductivity κ', pure isotope diamonds should have nearly the same thermal conductivity. However, the measured thermal conductivity of 99% ^{13}C diamond is significantly lower than that of ^{12}C diamond. It will be attributed to the 1% isotope impurity which decreases the phonon mean free time. According to the past research (Ref. [5]), even 1% impurity effect thermal conductivity dramatically.

The measured thermal conductivity of superlattices is lower than that of pure isotope diamonds, which fits the simulation result. The long-period superlattice structure reduces the thermal conductivity. The longer the superlattice period, the more folds in the Brillouin zone, the greater the drop in phonon group velocity near the folded Brillouin zone (Fig. 2).

Figure 3 illustrates the effect of different sublattice layer number (n_{12C},n_{13C}). (4,2) is higher than (6,2) because of the low period. The κ' value of (6,2) is higher than (2,6) because of the high ratio of ^{12}C. However, it is interesting that (4,4) is between (2,6) and (6,2) despite the ^{12}C ratio is between the two. Furthermore, we use (n_{12C},2) and (2,n_{13C}) for discussing the influence of impurity on thermal conduction as shown in Fig 4. The significant reduction of κ' in (n_{12C},2) and (2,n_{13C}) (~50%) is due to the destruction of the periodic structure of the lattice. These can explain the significant drop in thermal conductivity containing impurities.

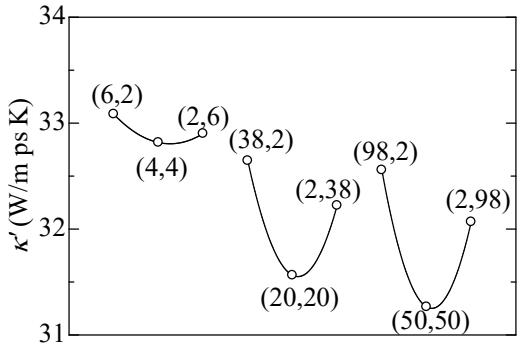

Fig. 3 Lattice thermal conductivity of asymmetry superlattice diamonds. Solid curves are the guides.

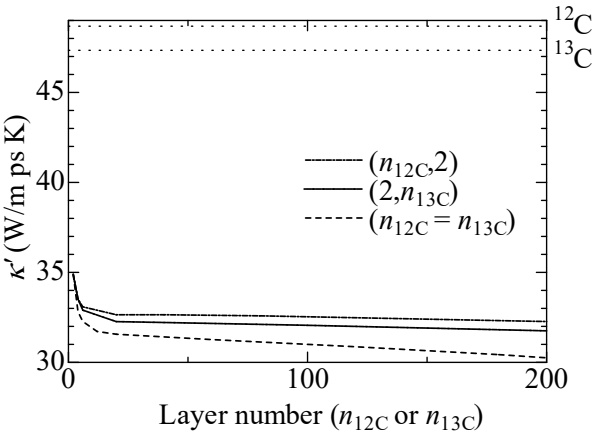

Fig. 4 Lattice thermal conductivity of (n_{12C},2), (2,n_{13C}), and (n_{12C},n_{13C}).

Acknowledgment

This work was supported by KAKENHI Grant-in-Aid for Scientific Research A (JP19H00862).

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