



Coupled theoretical interpretation and experimental investigation of the anisotropy of the lattice thermal conductivity of Bi_2Te_3 single crystal

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ABSTRACT

The Debye model is modified for the calculation of the lattice thermal conductivity and used to gain insight into the anisotropy of Bi_2Te_3 . In this work, the Debye temperature is not used to estimate the cutoff frequencies of the phonons that carry heat. The cutoff frequencies are defined by setting an upper limit to the energy of acoustic phonons using the complete dispersion relations. The anisotropy of the thermal conductivity is found to be unrelated to the anisotropy of the sound velocities. It is found that the sound velocity is almost isotropic when the longitudinal and two transversal waves are added together. In addition the relaxation time must be a function of the cutoff frequencies and counterbalances the anisotropy arising from the variation of the number of acoustic phonons traveling in various directions. It is concluded that the anisotropy of the thermal conductivity is mostly related to the Grüneisen's constant.

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1. Introduction

The kinetic theory of the lattice thermal conductivity has provided only qualitative information so far. Measurement of the thermal conductivity as a function of the temperature was the only tool used to check the theory but there were so many so-called adjustable parameters in the theory that an agreement between theory and experiment was almost always found. Significant conceptual differences in the theory could not be checked experimentally. As a result the kinetic theory has not been significantly improved since the 1960s [1,2].

The key idea presented here is to study the anisotropy of the thermal conductivity theoretically and compare the prediction obtained with published experimental data for Bi_2Te_3 [3]. What did motivate us is to give a simple but not simplistic explanation for the anisotropy of the lattice thermal conductivity of Bi_2Te_3 . This article about the lattice thermal conductivity complements our article about the anisotropy of the electrical properties of Bi_2Te_3 [4].

In the present article realistic dispersion relations of Bi_2Te_3 are used to define the cutoff frequencies of acoustic phonons along the principal axes. The speed velocity in anisotropic medium are calculated exactly using the Christoffel's equation [5]. It is shown that the cutoff frequencies must also enter in the calculation of the relaxation time in order to explain the anisotropy of the lattice thermal conductivity of Bi_2Te_3 .

2. Cutoff-frequencies

There are no very well established methods to extract the cutoff frequencies of the acoustic phonons from the complete dispersion relation. According to Han and Klemens, the cutoff frequencies are the lowest frequencies mode at the zone boundary [6]. This definition is also adopted by Morelli, Heremans and Slack [7]. In this work the cutoff frequencies of the acoustic phonons are defined where an acoustic branch crosses an optical branch. This choice is justified later. The value of the cutoff frequencies so defined are (Fig. 1) $\theta_z = 51$ K (along the $\vec{c}$ axis), $\theta_x = 69$ K and $\theta_y = 65$ K (perpendicular to the $\vec{c}$ axis). The cutoff-frequencies are clearly anisotropic. Since the cutoff frequency is the lowest along the $\vec{c}$ axis, it is tempting to explain the lowest thermal conductivity in that direction with the concept of cutoff frequency of the acoustic phonons. Nevertheless results that are presented later in this article cast a shadow over this idea. Still the cutoff-frequencies of the acoustic phonons are 2–3 times smaller than the frequency obtained from the Debye temperature and this already explains why the thermal conductivity of Bi_2Te_3 is low. A cutoff frequency about 76 K was estimated in Ref. [8].

3. Sound velocity in anisotropic medium

The sound velocity in Bi_2Te_3 has been calculated using the Christoffel's equation in every direction [10]. Examples are shown in Fig. 2 for the speed velocity calculated in the basal plane of the hexagonal unit cell and perpendicular to it. The detail of the

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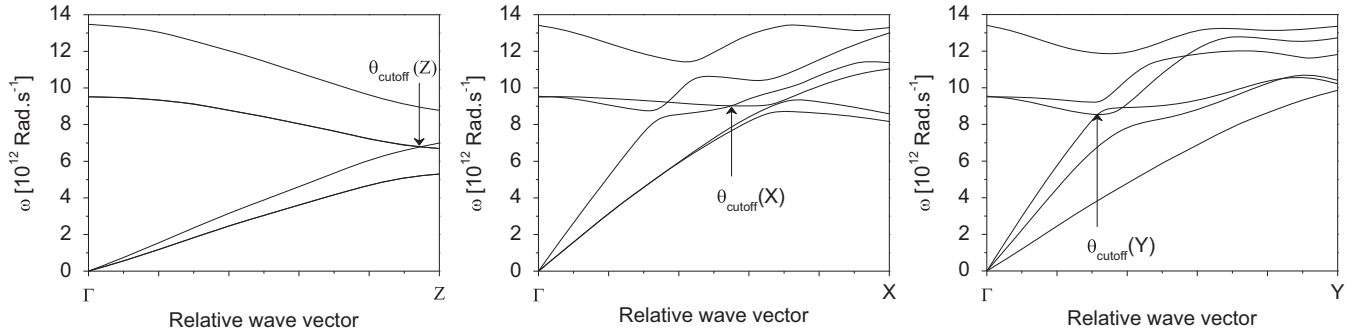


Fig. 1. Dispersion relation of Bi_2Te_3 along the principal axes taken from Ref. [9]. The cutoff frequencies in this work are defined exactly where the acoustic branch cross the optical branch. Only the first three optical branches are shown.

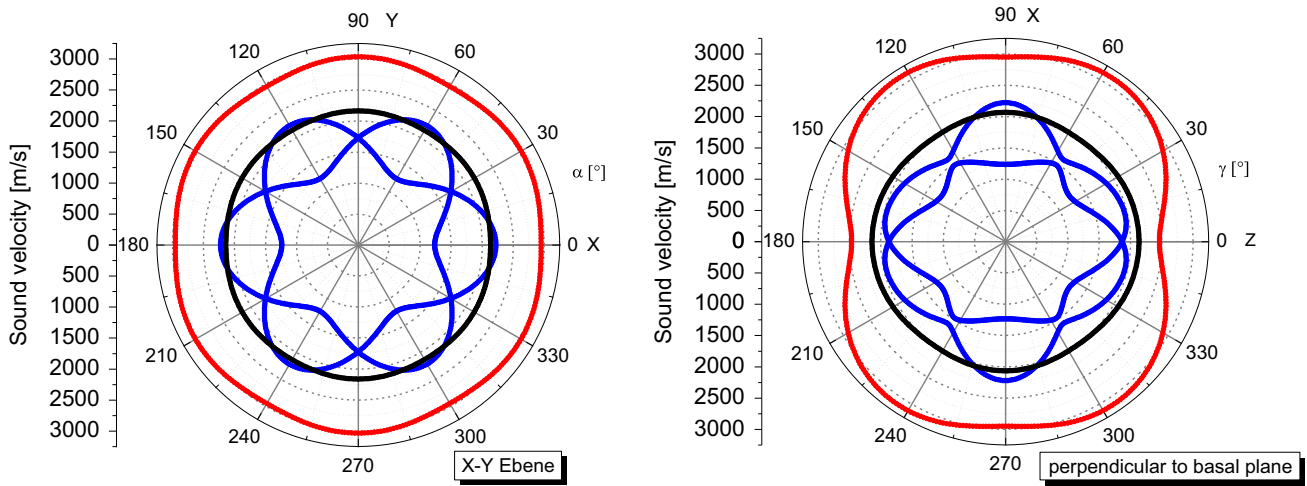


Fig. 2. Sound velocity of Bi_2Te_3 in the basal plane of the hexagonal unit cell and perpendicular to it. The longitudinal polarization and transversal polarizations are shown in red and blue, respectively. The average of the sound velocity of the longitudinal and two transverse waves is black. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

calculation can be found in Ref. [11]. The main result is that the anisotropy of the lattice thermal conductivity measured in Bi_2Te_3 is not explained by the anisotropy of the sound velocities.

4. Relaxation rate

The scope of this section is limited to justify at least partially the most common expression for the relaxation rate due to the Umklapp-Processes: [12,13]

$$\tau^{-1}(\omega) = B\omega^2 \frac{T}{\theta_D} e^{-\theta_D/bT} \quad (1)$$

where

$$B = \frac{\hbar\gamma^2}{Mv^2} \quad (2)$$

θ_D is the Debye temperature, M is an average mass of a single atom in the solid, v is the average sound velocity and γ is the Grüneisen's constant. b is an adjustable parameter.

A general and meaningful formulation of the relaxation rate starts with a description of the collisions that are to our point of view the most likely to happen. A phonon identified by (') hits another phonon (") so that it is reflected back. The incoming and reflected waves tend to build standing waves similar to an optical wave with no net momentum (acoustic phonon '''). Since phonons are waves, and waves can be viewed as medium of periodic

impedance (period a), the scattering satisfies Bragg conditions:

$$\lambda = 2a/n \quad (3)$$

If $n > 1$, energy is transferred to phonon modes of higher energy. Since the energy is conserved, the energy of the incoming and reflected wave will decrease [11,14] and at the end there will be only optical phonons remaining with no net momentum. The time needed for phonons to go through the above described process may be called the relaxation time. In this work, the cutoff frequencies of the acoustic phonons are defined exactly where an acoustic branch crosses an optical branch because the transition probabilities $\sim (1 - \cos(\Delta E/E))/(\Delta E/E)^2$ between two states of energy E decreases sharply when the energy of the two states moved of ΔE apart [2].

In order to calculate the relaxation time τ , we consider a phonon moving with the speed v scattered when its path crosses the cross-sectional area S of a scattering center (a phonon). Since τ is the mean time taken for one scattering processes to occur, the mean free path l between scattering processes is $v\tau$. If N is the density per unit of volume of target-phonons, then in the volume Sl , there is one scattering center. Therefore $(Sv\tau)N = 1$. Thus the relaxation rate is given by:

$$\tau^{-1} = SvN \quad (4)$$

It is evident that N is a function of the cutoff frequencies since scattering occurs when the transition probability is the highest,

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