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Evidence of ballistic thermal transport in lithium niobate at room temperature

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ABSTRACT

In ballistic transport, heat carriers such as phonons travel through the solid without any scattering or interaction. Therefore, there is no temperature gradient in the solid, suggesting zero thermal conductivity by Fourier's law. Ballistic transport is typically seen in high purity crystals at either temperatures below ~10 K, or physical size below ~100 nm, where the mean free path of the carrier is larger than the solid itself. In this letter, we show evidence of ballistic transport at room temperature in lithium niobate wafers in the in-plane and cross-plane directions under both steady state and high frequency heating that are monitored using both infrared and resistance thermometry. We report phonon mean free path in lithium niobate around 425 μm, which is about 50 times higher than the largest phonon mean free path in the literature at room temperature.

1. Introduction

In dielectric and semiconducting solids, heat is mainly conducted by phonons, which interact with other phonons, defects and interfaces. The average distance they travel between each scattering event is called the mean free path, which is typically on the order of ~100 nm. Phonon transport in these solids has three possible mechanisms: a) by ballistic transport, where the phonons travel through the solid without any scattering, b) by second sound, where the energy transport is a wave like phenomenon and the phonon momentum is conserved during most of the scattering process and c) by diffusion, which is observed in almost all the bulk materials. The ballistic transport and second sound are observed at very low temperatures and in materials of high purity. These cases are drastically different from classical Fourier conduction, which erroneously suggests no heat transfer because there is no temperature gradient. When the characteristic size of the solid is much larger than the mean free path of the phonon, there is massive scattering and the heat flow is diffusive. In reality, the material appears to transfer the heat without getting heated itself. Ballistic heat transfer is seen in nano-systems where the solid size is lesser or comparable to the phonon mean free path at room temperature [1–3]. Two-dimensional materials like graphene [4,5] also shows such behavior. To date, the largest phonon mean free path at room temperature is reported to be around 8.3 μm in SiGe nanowires [6].

Fourier's law of heat conduction, given by Eq. (1), assumes local thermal equilibrium and the heat flow to be diffusive in nature. In a solid with thermal conductivity κ, it defines the heat flux vector **q** is proportional to the temperature gradient ∇T in the material. Derived from the kinetic theory, the thermal conductivity is related to other physical properties as in Eq. (2).

$$\mathbf{q} = -\kappa \nabla T \tag{1}$$

$$\kappa = \frac{1}{3} C v \lambda \tag{2}$$

where, C is the specific heat per unit volume of the material, v is the speed of sound in the material and λ is the phonon mean free path. In the case of ballistic phonon transport, the Fourier law breaks down and is known to over-predict the thermal transport [7,8]. In such cases, the Boltzmann Transport Equation (BTE), given below by Eq. (3), has to be solved to study the thermal transport.

$$\frac{\partial f}{\partial t} = \mathbf{v_g}.\nabla f + \frac{\partial \mathbf{k}}{\partial t}.\nabla_k f = \left|\frac{\partial f}{\partial t}\right|_{collision} \tag{3}$$

where, f is the phonon distribution function dependent on position vector **r**, time t, wave vector **k** of the phonons. $\mathbf{v_g}$ is the group velocity vector of phonons which is defined as

$$\mathbf{v_g} = \frac{\partial \omega}{\partial \mathbf{k}} \tag{4}$$

where, ω is the phonon frequency. The right hand side of Eq. (3) denotes the change in the phonon distribution due to the collisions. There are several approaches to solve the BTE using various approximations since a direct analytical solution to its raw form is not practical. Few of

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Nomenclature

q	Heat flux vector
T	Local temperature
v_g	Phonon group velocity
ω	Phonon frequency
h	Natural heat transfer coefficient
T_∞	Ambient temperature
P	Specimen perimeter
y	distance perpendicular to specimen length
x	distance along the specimen length
κ	Thermal conductivity
C	Specific heat
λ	Phonon mean free path
v	Speed of sound
f	Phonon distribution function

them include the relaxation time approximation [9], neglecting the optical phonons, isotopic approximation [10] and gray approximation where the relaxation time and phonon velocity are taken to be frequency independent. Literature exists on computational studies of thermal transport in ballistic regime [2,11–14]. It is widely accepted that phonons have majority states with mean free paths in the order of ~100 nm. Despite that, a minority of phonon states exist with mean free path as big as 10 μm at room temperature and still make a strong contribution to thermal conductivity of the solid [15].

2. Experimental design

There is a very little body of experimental evidence of long mean free paths at the *meso* or macro scales at room temperature. Nanostructures and cryogenic temperatures have a limited utility to applications. In this communication, we present the experimental observation of ballistic phonon transport at room temperature in a bulk lithium niobate wafer. Lithium niobate is a dielectric material with attractive piezoelectric, optical, electro-optic and photorefractive properties. It has non-linear optical properties which makes it an encouraging material for conversion of thermal radiation [16]. It is also cheaply available with a very high Curie temperature (~1500 K) [17] that makes it favorable for applications in high temperature devices. To demonstrate ballistic thermal transport at room temperature, we performed steady state thermal transport experiments along in-plane and cross-plane directions, followed by high frequency heat pulse experiments typically used to capture non-diffusive transport. We then estimated the phonon mean free path by measuring thermal conductivity as function of specimen length. We also performed finite element simulation to highlight the drastic difference between our experimental results and the conventional Fourier transport.

3. Experimental results and discussion

3.1. In-plane ballistic transport

To map the in-plane thermal transport, we patterned 30 μm wide nickel lines in serpentine shape to act as a thin film heater. This is shown in Fig. 1a. Next to this heater, several thin nickel film lines were patterned in parallel so that resistance thermometry can be used to measure the local temperature for cross-validation of the infrared thermometry. Fig. 1a shows the infrared (IR) image after passing current through the thin film heater. This excites the phonons in that region that propagate until they scatter at a boundary such as the heat-sink films. To study the experimentally obtained temperature domain, we developed a multi-physics model incorporating Joule heating and diffusive heat transfer using the same geometry and heating current input to the commercially available COMSOL software. In the simulation, we assumed the interfacial thermal resistance to be 7.7×10^{-08} [m^2 K/W], a value intentionally setup to be slightly higher than typical in order to completely eliminate the possibility that the results are artifacts from thermal resistance. The simulation results are shown in Fig. 1b. The stark contrast between the two temperature profiles is highlighted in Fig. 1c, which shows the temperature line scan along x-x′ direction. It shows that diffusive heat transfer would be localized in the heater area, gradually decreasing to the room temperature. Given the low thermal conductivity (~4 W/m-K) of the lithium niobate substrate, very little heat is expected to be transported to the heat sinks, indicated by the clear absence of any temperature peaks on the metal heat-sink lines. However, the IR images capture a completely different physics, where the heater lines show high temperature spikes but the substrate between two heater lines do not heat up appreciably. To highlight that substrate carries the heat without itself getting hot, we locate four locations on the substrate marked as A, B, C and D in Fig. 1c. Intriguingly, the substrate temperature is close to room temperature right next to the heater (point A) and it remains fairly constant through the points B, C and D. However, very distinct temperature peaks are seen in the metal heat sink lines next to these locations. These observations clearly show that the phonons propagate in a ballistic manner, i.e., they do not create temperature gradient unless they scatter at the boundary (which is the interface between metal and the substrate).

3.2. Cross-plane ballistic transport

To demonstrate cross-plane ballistic transport, we mount a 2 mm × 2 mm ceramic heater on one side of the lithium niobate wafer and observe the temperature profile along the thickness direction. Fig. 2a shows a cross-sectional image from IR microscope, which shows a sharp temperature drop at the interface after which the temperature profile is uniform inside the lithium niobate. This is better seen in Fig. 2b that shows the line scan results on the temperature. Once again, the lithium niobate wafer carried the heat without getting hot; however, a small temperature spike (marked by the oval) was seen at the boundary of the solid. This is due to energy dissipation when the phonons scatter at the substrate boundary, which reconfirms the ballistic transport. Fig. 2c shows the simulation results from COMSOL for the same operating and boundary conditions. Here, the diffusive heat transfer develops a small temperature gradient along the thickness direction. This is because the thin glass substrate heats up due to phonon scattering, which is absent in the experimental observations (Fig. 2b).

To revalidate the signature of ballistic transport, we modified this cross-plane experiment by attaching a second material (a Kapton foil) to the lithium niobate. This is shown in Fig. 2d. If there zero thermal gradient in the material were a result of extremely high thermal resistance (or no heat flow in the material,) then one would expect the second material (kapton) to remain at room temperature. However, this was not the experimental observation, where the niobate substrate carried the heat to the Kapton foil to raise its temperature to nearly the magnitude of the heater itself, while it remained at a lower temperature.

3.3. Transient heat pulse response

Classical studies [18,19] on ballistic transport have used transient heat pulse on one side of the solid while capturing the thermal response on the other side as function of time using a thin film bolometer. Fig. 3a shows the landmark results on highly pure NaF crystals at cryogenic temperatures [20], where two distinct response peaks appear. The first peak (marked as L) has smaller amplitude and originates from longitudinal phonons while the significantly higher second peak (marked as T) is originated from transverse phonons. At room temperature, the peaks flatten dramatically to a single waveform. To capture this