Phonon-polariton in heat conduction

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abstract

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I. INTRODUCTION

It is well-known that the contribution of radiative transport to thermoconductivity of majority of solids is important only at rather high temperatures of the order of a few thousands K. At much lower temperatures the radiative transport is usually small because is small the density of photon states which have an energy of the order of $k_B T$. The energy of the transverse photons, responsible for the radiative transport (we assume that the medium is isotropic), is $E(k) = \hbar ck/\sqrt{\varepsilon}$, where c is the velocity of photon in vacuum, k is its wave vector and ε is the dielectric constant of medium. The density of states is proportional to $k^2(\frac{dk}{d\omega})$ or (for transverse photons) to ω^2 what is a smooth function of ω and small for small ω.

The situation changes if to take into account the dependence of dielectric constant on ω which may be strong in the region of dipole allowed resonances. In this region of spectrum the interaction of dipole active quasi-particles (transverse optical phonons) with transverse photons (retardation effect) is responsible for the appearance of a new quasi-particles, so called phonon-polaritons [1,2]. For these quasi-particles most important is vicinity of transverse optical phonon frequency where dielectric constant has a resonance. The polariton dispersion in the region of isolated resonance (a dependence of its frequency on wave vector) can be found from the relation

$$\frac{k^2 c^2}{\omega^2} = \varepsilon(\omega), \tag{1}$$

$$\varepsilon(\omega) = \varepsilon_b \frac{\omega^2 - \omega_\parallel^2}{\omega^2 - \omega_\perp^2}, \tag{2}$$

where $\omega_\parallel$ and $\omega_\perp$ are frequencies of longitudinal and transverse optical phonons. For some crystals the transverse-longitudinal splitting $\Delta = \omega_\parallel - \omega_\perp$, which is proportional to the oscillator strength at the resonance frequency $\omega_\perp$, can be rather large. For example, for SiC crystal[4,5] $\omega_\perp$=793cm^{-1}, $\omega_\parallel$=969 cm^{-1} and, thus, Δ=176cm^{-1}, for crystal MgO[6], where $\omega_\perp$=396cm^{-1}, $\omega_\parallel$=719cm^{-1} this splitting is even larger: Δ=323cm^{-1}

If we take into account the dissipation or scattering of polaritons we can use for dielectric constant a more

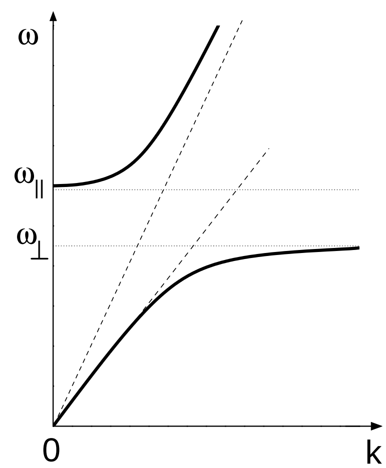

FIG. 1:

general expression

$$\varepsilon(\omega) = \varepsilon_b \frac{\omega^2 - \omega_\parallel^2}{\omega^2 - \omega_\perp^2 - 2i\gamma\omega}, \tag{3}$$

The approach to calculate the polariton heat conductivity is dependent on the relation between size of sample and the length of polariton mean-free-path. If this length is larger than the sample size it is necessary to consider polaritons in ballistical regime. If, however, the size of sample is large in comparison with the length of polariton mean-free -path we can use the same statistical random walks approach which usually is in use in calculation of phonon heat conductivity in solids. In this note below we consider crystals MgO and SiC at temperature

$T \approx 1000K$. At this temperature the states of polaritons with energy $E \approx 0.1eV$ are mainly populated and statistical approach in calculation of polaritons heat conductivity can be justified because for these phonon-polaritons in mentioned crystals a mean-free- path is rather small. For example, as it follows from the measurements of absorption in MgO crystal[3], the absorption coefficient at temperature $T \approx 1000K$ changes in wide interval values up to $10^5 cm^{-1}$ but in all cases it is larger than $10^2 cm^{-1}$ at least in the interval of wave numbers $150 - 1500cm^{-1}$ (unfortunately, we have no other measurements). It means that a polariton mean-free-pathΛ for these wave numbers is less than value of order of $\approx \frac{1}{150} \approx 0.07cm$ and similar situation we meet for

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many another crystals as it follows from experimental data on light absorption. It means that for calculation of polariton thermoconductivity of the sample with thickness of order of 1cm we can use statistical theory taking into account the contribution to thermoconductivity the phonon-polaritons with the mean-free-path smaller than the sample size. We will show that this restriction is important for the temperature interval where the statistical theory can be used. The total thermoconductivity is the sum, roughly speaking, of two parts arising from ballistical and diffusive propagation of polaritons. Thus, in comparison with experimental data it is necessary to take into account that the part of total thermoconductivity arising from statistical approach can determine only the lower limit of its total value.

II. THERMAL CONDUCTIVITY

The thermal conductivity $\kappa(T)$ can be calculated by the using of following well-known expression

$$\kappa(T) = \frac{1}{3}\sum_p \int C(\omega)v(\omega)\Lambda(\omega)d\omega, \tag{4}$$

where ω is the polariton frequency, $C(\omega)$ is its thermal capacity , $v(\omega)$ is its the group velocity, and $\Lambda(\omega)$ is its mean-free-path. The sum is carried out over two transverse polariton polarizations p.

In order to determine $\kappa(T)$ we firstly have obtained C, v, and Λ. The polaritons energy at thermal equilibrium can be written as

$$E(\omega, T) = \hbar\omega \frac{D(\omega)}{\exp(\hbar\omega/k_BT) - 1}, \tag{5}$$

where the density of states $D(\omega)$ is given by

$$\frac{D(\omega)}{V} = \frac{4\pi k^2}{(2\pi)^3}\frac{dk}{d\omega}. \tag{6}$$

Therefore, the thermal capacity can be written as

$$\begin{aligned} C(\omega) &= \frac{1}{V}\frac{dE}{dT} = \\ &= \frac{D(\omega)}{V}\frac{(\hbar\omega)^2 e^{\hbar\omega/k_BT}}{k_BT^2(e^{\hbar\omega/k_BT}-1)^2}. \end{aligned} \tag{7}$$

As the absorption is rather weak we can express the group velocity as

$$v(\omega) = \frac{d\omega}{dk}. \tag{8}$$

The last quantity to determine is the mean free path. Since the intensity I is proportional to the squared electrical field we have

$$I \sim |E|^2 \sim e^{i2kz} = e^{i2(n'+in'')\omega z/c} \sim e^{-2n''\omega/c} = e^{-z/\Lambda(\omega)} \tag{9}$$

thus,

$$\Lambda(\omega) = \frac{c}{2\omega n''(\omega)}. \tag{10}$$

Using the relation

$$\frac{k^2(\omega)c^2}{\omega^2} = (n' + in'')^2 = \varepsilon(\omega) = \varepsilon' + i\varepsilon'', \tag{11}$$

and assuming weak absorption ($(n'')^2 \simeq 0$) one can obtain

$$\varepsilon'(\omega) = \varepsilon_\infty \left(1 + \frac{(\omega_\parallel^2 - \omega_\perp^2)(\omega_\perp^2 - \omega^2)}{(\omega_\perp^2 - \omega^2)^2 + 4\Gamma^2\omega^2}\right), \tag{12}$$

$$\varepsilon''(\omega) = \varepsilon_\infty \left(\frac{2\Gamma\omega(\omega_\parallel^2 - \omega_\perp^2)}{(\omega_\perp^2 - \omega^2)^2 + 4\Gamma^2\omega^2}\right), \tag{13}$$

$$n'(\omega) = \sqrt{\varepsilon'(\omega)}, \qquad n''(\omega) = \frac{\varepsilon''(\omega)}{2n'(\omega)}. \tag{14}$$

In order to check the approximation considered here for the calculation of $\Lambda(\omega)$ we plot in Fig. 1 the absorbance obtained from (11) and experimental values for the MgO crystal. We can notice a reasonable agreement between them.

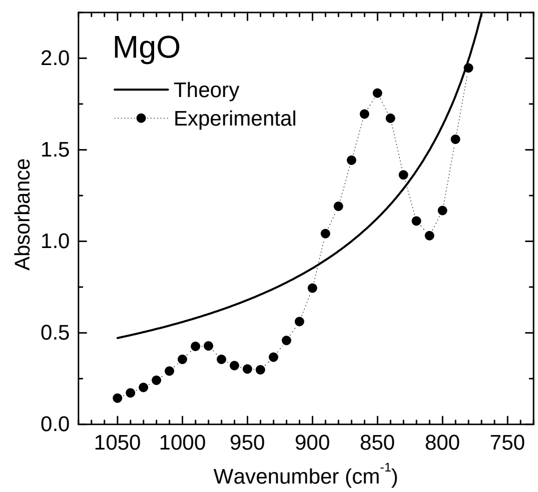

FIG. 2: Absorption spectra of MgO. Experimental points correspond to the MgO crystal at T=305 K with 0.16 mm thick[7]. The dotted line just connects the points.

Grouping (8), (9), (11), and (13)-(15) and summing over two photon polarizations, the thermal conductivity, considering polaritonic resonance, becomes

$$\kappa(T) = \frac{k_B^3T^2}{3\pi^2\hbar^2c}\left[\int_0^{x_\perp} h(x)dx + \int_{x_\parallel}^{\infty} h(x)dx\right] \tag{15}$$

where

$$x(T) \equiv \frac{\hbar\omega}{k_BT}, \;\; x_\perp(T) \equiv \frac{\hbar\omega_\perp}{k_BT}, \;\; x_\parallel(T) \equiv \frac{\hbar\omega_\parallel}{k_BT}, \tag{16}$$

and

$$h(x) \equiv \frac{x^3 e^x}{(e^x-1)^2} \frac{\sqrt[3]{\varepsilon'(x)}}{\varepsilon''(x)} \tag{17}$$

III. RESULTS AND DISCUSSIONS

We have used data for SiC and MgO in order to establish a comparison with the thermal conductivity values obtained here. The experimental data considered were: $\varepsilon_b = 6.7$, $\omega_\perp$=793 cm^{-1}, $\omega_\parallel$=969 cm^{-1}, and Γ=4.76 cm^{-1} for SiC[4,5], and $\varepsilon_b = 2.96$, $\omega_\perp$=396 cm^{-1}, $\omega_\parallel$=719 cm^{-1}, and Γ=7.60 cm^{-1} for MgO[6]. Experimental values of thermal conductivity were extracted from[8] for SiC (p. 279) and for MgO (p. 283).

Figure 2 presents the thermal conductivity as function of temperature considering one polaritonic resonance. Experimental data are also shown. For $T > 300$K, $\kappa(T)$ behaves as $\kappa(T) \sim T^5$.

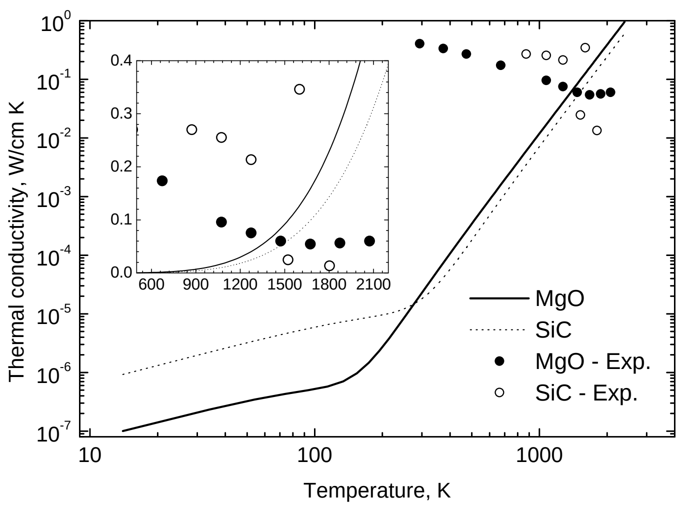

FIG. 3: Thermal conductivity as function of temperature. The lines were obtained using the expression (16). The inset graph is the zoom for the region 600 K< T <2200 K.

IV. CONCLUSIONS

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