Analysis of heat transport in the iron oxyarsenide TbFeAsO$_{0.85}$.

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Abstract. Thermal conductivity $\kappa(T)$ of a high-quality polycrystalline TbFeAsO$_{0.85}$ sample is theoretically investigated. The lattice contribution to the thermal conductivity ($\kappa_{\text{ph}}$) is discussed within the Debye-type relaxation rate approximation in terms of the acoustic phonon frequency and relaxation time below superconducting transition temperature ($T_c = 42.5$ K). The theory is formulated when heat transfer is limited by the scattering of phonons from defects, grain boundaries, charge carriers, and phonons. The $\kappa_{\text{ph}}$ dominates in TbFeAsO$_{0.85}$ and is an artifact of strong phonon-impurity and -phonon scattering mechanism. Our result indicates that the maximum contribution comes from phonon scatters and various thermal scattering mechanisms provide a reasonable explanation for maximum appeared in $\kappa(T)$.

1. Introduction

The recent discovery of high superconducting transition temperatures in FeAs based compounds has generated great interest in the scientific community [1], and because the strong interplay between the structural, magnetic and superconducting properties has led to close comparisons with the physics of the superconducting copper oxides [2, 3]. It has been also recognized that FeAs-based compounds can be promising thermoelectric materials in refrigeration applications around liquid nitrogen temperatures [4]. These aspects have generated several motivating studies in this direction by replacing O atoms by F atoms at a concentration of 3-13 atoms%, with enhancing of critical temperature from 26 K to highest of about 56 K [5-7]. These quaternary superconductors have a rather simple structure of alternating [Fe$_2$As$_2$]$^{2-}$ and [Ln$_2$O$_2$]$^{2-}$ layers with eight atoms in a tetragonal unit cell (P4/nmm) [8], where the [Fe-As]$_2$- layer is thought to be responsible for superconductivity, while the [Ln$_2$O$_2$]$_2$- layer provides charge carriers, resemblance to that of high-$T_c$ cuprates.

Very recently, the superconductivity has been realized by producing oxygen vacancies instead of F atoms doping [9 - 10], which can create more carriers in the charge reservoir layer, as considered necessary to tune the physical properties. Thus, the stacking of these layers and ionic states of constituent ions have decisive role in the transport and superconducting properties. Evolution of structural, electronic and thermal properties would be an important step toward realizing the potential technological scenario apart from richness of physics of this material class. Previously, we have reported the synthesis of highly pure single phase of TbFeAsO$_{0.85}$, x-ray absorption spectroscopy, and
anomalous thermal transport properties [11]. The electrical (with $T_c = 42$ K) and magnetic properties of the polycrystalline TbFeAsO$_{0.85}$ sample have been described elsewhere [10]. In this paper, we report the theoretical investigation of thermal conductivity $\kappa(T)$ of a high-quality polycrystalline TbFeAsO$_{0.85}$ sample experimental data. The $\kappa(T)$ measurement shows an anomaly in the vicinity $T_c$. The $\kappa_{ph}$ is discussed within the Debye-type relaxation rate approximation in terms of the acoustic phonon frequency and relaxation time below transition temperature. Our result indicates that the maximum contribution comes from phonon scatters and various thermal scattering mechanisms provide a reasonable explanation for maximum appeared in $\kappa(T)$.

2. The Model
We start with a model Hamiltonian that follows [12]

$$
H = \sum_k \varepsilon_k a_k^+ a_k + \sum_q \varphi(k_1,k_2) a_{k_1}^+ a_{k_2} + D \sum_{k,q} \left[ \frac{\hbar}{2\rho \omega_q} \right]^{1/2} a_{k+q}^+ a_k (b_q + b_{-q}) + \frac{R}{2N} \sum_{q_1,q_2} e^{i(q_1+q_2)\cdot r_i} \left[ \frac{\hbar \omega_{q_1} \hbar \omega_{q_2}}{4} \right]^{1/2} (b_{q_1} - b_{-q_1}^+)(b_{q_2} - b_{-q_2}^+) + H_{ph-ph}.
$$

Here, initial two terms is holes as carriers and phonon excitation. The third and fourth terms represent the carrier-impurity interactions and carrier-phonon interactions, respectively. The fifth term denote the phonon-impurity interactions and the last term stands for the phonon-phonon interaction. The notations $a$ ($a^+$) and $b$ ($b^+$) are the creation (annihilation) operators for holes and phonons. Further, $\varepsilon_k$ is hole free energy, phonon frequency of a wave vector $q$ is $\omega_q$, $D$ is the deformation-potential constant. $R$ is the relative ionic-mass difference $[(M''-M)/M''], \rho$ is the mass density of ions and $r_i$ stands for the position of defects. We begin with phonon contribution to thermal conduction. The thermal conductivity can be calculated from the Kubo formula [12]. It has contributions from both the phonons and the carriers. We first look for the lattice part and in the continuum approximation it follows

$$
\kappa_{ph} = \frac{k_B \hbar^2}{2\pi^2 v_s \omega_D} \int d\omega \omega^2 \tau(\omega)(\beta \omega)^2 \frac{e^{\beta \omega}}{(e^{\beta \omega} - 1)^2},
$$

with $k_B$ is the Boltzman constant, $v_s$ is the sound velocity, $\omega_D$ is the Debye frequency and $\beta = \hbar/k_B T$. The relaxation time is proportional to the imaginary part of the phonon self-energy. In the weak interaction case, it has been calculated to the lowest order of the various interactions. The relaxation times are expressed as

$$
1/\tau(\omega) = 2\text{Im} P(\omega/v_s, \omega) = 1/\tau_{ph-d} + 1/\tau_{ph-e} + 1/\tau_{ph-gh} + 1/\tau_{ph-ph},
$$

with the various relaxation times are defined as

$$
\tau_{ph-d}^{-1}(\omega) = (A/k_B^3)\omega^4 \hbar^3,
$$

$$
\tau_{ph-e}^{-1}(\omega) = B\omega n_F,
$$

$$
\tau_{ph-gh}(\omega) = v_s / L,
$$

$$
\tau_{ph-ph}^{-1}(\omega) = C(T\omega/h k_B^3).
$$
where $L$ is the sample size, $n_F$ is the Fermi-Dirac distribution function and $\Delta$ is the gap function. The notation $\tau_{ph-d}$, $\tau_{ph-c}$, and $\tau_{ph-gb}$ are the phonon scattering relaxation time due to defects, carriers, grain boundaries, and phonon-phonon interactions respectively. We note that to this order Mathiessen’s rule holds namely, that the inverse of the total relaxation time is the sum of the various contributions for the different scattering channels.

Further we define $A = \left[ \frac{3n_iR^2}{4\theta_i^2} \right]$ and $B = \frac{9\pi}{4} \left[ \frac{m}{3M} \right]^{3/2} \frac{D_P^2}{E_F^2}$, are parameters, which characterize the strengths of the phonon-defect and phonon-carrier, scattering process. Here, $n_i$ is the density of impurities or defects, $E_F$ is the Fermi energy of holes as carriers, and $m$ is its mass.

### 3. Results and Discussion

The experimental results of the thermal conductivity on polycrystalline TbFeAsO$_{0.85}$ used in this work were reported previously [11] and is shown in Fig. 1 for comparison. At low temperatures, $\kappa$ increases with temperature and a broad peak appear around 27 K and then a steep fall as temperature increases further. The occurrence of such a low temperature maximum is a typical feature of solids due to the reduction of thermal scattering, i.e., the phonon mean free path is approximately equal to the crystal site distance and ascribed to the generalized Umklapp process at low temperatures. Further, $\kappa$ is found to be almost saturating as the temperature increases above 75 K. In order to elucidate these distinguishing features of the TbFeAsO$_{0.85}$ polycrystalline sample, we have analyzed our data in the framework of Kubo formula as described earlier. Here, we have neglected the contribution from electrons below $T_c$ because of the condensed superconducting pairs at low temperatures. In the calculation of temperature dependent thermal conduction of TbFeAsO$_{0.85}$, we use the parameters which characterize the strengths of the phonon-defect, phonon-electron, and phonon-phonon scattering process as $A = 3.9 \times 10^{-7}$ K$^{-3}$, $B = 0.12$ and $C = 0.5$ K$^{-6}$ sec$^{-1}$, respectively. The length of the sample is about 3 mm and $v_s = 4.76 \times 10^5$ cm sec$^{-1}$.

We first qualitatively discuss the properties of thermal conductivity due to phonons. Figure 2 shows our results for phonon thermal conductivity of TbFeAsO$_{0.85}$. As the temperature is lowered through $T_c$, the quasi particle excitation condensed into the ground state and they cannot scatter phonons. The phononic thermal conductivity, thus, increases exponentially as the temperature increases in the absence of the other scattering mechanism. Although the phonon thermal conductivity experiences an exponential increase below the transition temperature, the presence of the defect, and

**Figure 1.** Variation of thermal conductivity as a function of temperature.

**Figure 2.** Variation of Phononic thermal conductivity with temperature: Effect of various phonon scattering mechanism.
the electron scatterings set a limit on its growth, as a consequence the phononic thermal conductivity diminishes as the temperature increases. At much higher temperature phonon-phonon scattering becomes more effective and decreased mean free path of phonon is responsible for decrease in thermal conductivity at higher temperature. The maximum position depends on the relative magnitudes of the phonon-electron, phonon-defect and phonon-phonon scattering processes.

It is meaningful to comment that the contribution of the electron-impurity towards thermal conductivity to the phonon thermal conductivity is about 1% in the temperature domain \(0 < T < T_c\). Phonons are, then, the sole carriers of heat in this temperature domain. Further the all the contributions are clubbed together and temperature dependent of total thermal conductivity is shown in Figure 2 along with experimental data. The present analysis on thermal conductivity shows good agreement with the experiments results.

4. Conclusion

Thermal conductivity \(\kappa(T)\) of a high-quality polycrystalline TbFeAsO\(_{0.85}\) sample is investigated. The \(\kappa(T)\) measurement shows an anomaly in the vicinity of superconducting transition temperature \(T_c = 42.5\) K. The lattice contribution to the thermal conductivity \(\kappa_{\text{ph}}\) is discussed within the Debye-type relaxation rate approximation in terms of the acoustic phonon frequency and relaxation time. The theory is formulated when heat transfer is limited by the scattering of phonons from defects, grain boundaries, charge carriers, and phonons. Our result indicates that the maximum contribution comes from phonon scatters and various thermal scattering mechanisms provide a reasonable explanation for maximum appeared in \(\kappa(T)\). The numerical analysis of heat transfer below \(T_c\) shows similar results as revealed from experiments.

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