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Temperature dependence of thermal conductivity of vanadium substituted BPSCCO system between 10 and 150 K

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Abstract. **Thermal conductivity of a set of $(Bi_{0.8}Pb_{0.2-y}V_y)_2Sr_2Ca_2Cu_3O_{10+d}$ ($0 = y \leq 0.05$) pellets in the temperature range between 10 and 150 K is reported. Vanadium substitution influences strongly the magnitude of thermal conductivity (l) over the entire temperature range. But the nature of $l(T)$ dependence remains similar to that generally observed for HTSCs. The electronic contribution to the total l in the normal state is estimated to be ~ 25%. We have attempted to examine our data, assuming the role of both electrons and phonons in the origin of the $l(T)$ behaviour below T_c. Observed temperature variation of $l(T)$ for the present set of samples could be explained very well assuming this electron + approach. Some of the microscopic quantities estimated from the best-fit parameters give reasonable values.**

Keywords. **Thermal conductivity; high temperature superconductors; vanadium substitution; electron + phonon approach.**

1. Introduction

In recent years, thermal conductivity (l) of high temperature superconductors has been studied extensively (see reviews by Jezowski and Klamut 1990; Nunez-Regueiro and Castello 1991; Uher and Ginsberg 1992; Uher *et al* 1994) owing to its advantage in probing the scattering processes of phonons and the charge carriers in both normal and superconducting states. Such measurements also provide valuable information regarding the superconducting energy gap, electron–phonon coupling parameters, excitation spectrum and life times and order parameter symmetry (Ausloos and Houssa 1999). Characteristic features of $l(T)$ which are now known for La- (Bernasconi *et al* 1988), Y- (Uher and Kaiser 1987), Bi- (Peacor and Uher 1989) and Tl- (Aliev *et al* 1989; Uher *et al* 1991) based high T_c superconductors is: a rapid rise in l with increasing T below T_c which reaches a broad peak at $T = T_c/2$. It is then followed by a decrease and a sharp break at $T = T_c$. In sintered samples, thermal conductivity peak below T_c is nearly 20–25% higher than the l value at T_c, which for single crystals has been reported to be nearly 100%. In order to explain the observed nature of heat transport in high T_c superconductors, several theoretical models and propositions have been reported so far. Yet there still exists lack of consensus in regards to the origin of the features in $l(T)$, particularly for $T < T_c$. There are primarily two alternative interpretations of $l(T)$ behaviour of ceramic superconductors. One (Tewordt and Wolkhaussen 1989, 1990) attributes it to the decrease in the phonon scattering by carriers due to the condensation of electrons into non-phonon scattering Cooper pairs below T_c. The other explanation is based on purely electronic contribution (Yu *et al* 1992; Ausloos and Houssa 1993) and owes its origin to a decrease in the quasiparticle scattering rate below T_c. It may be noted that supporting data for both the models are rather extensive (Tewordt and Wolkhaussen 1989; Peacor *et al* 1991; Cohn *et al* 1992; Ting *et al* 1994; Houssa and Ausloos 1996a; Matsukawa *et al* 1996, 1999; Schachinger and Carbotte 1998; Verebelyi *et al* 1999). Considering the thermal conductivity peak below T_c to have contributions from both phonon and electrons, Castellazzi *et al* (1997) recently explained extremely well their $l(T)$ data on *c*-axis oriented BSCCO–2223 tape. However, the validity of this new approach has not been examined yet for any type of sintered polycrystalline high T_c superconductors.

It has been reported (Fung *et al* 1990; Xin *et al* 1990) that double substitution of V and Pb in Bi–Sr–Ca–Cu–O leads to a balance of atomic size and coulombic forces in the atomic arrangement within a superconducting unit cell and that the double substitution represents an ordered process preserving the 2223 structure. Thermal conductivity of such a system has not yet been studied in detail. In the present communication, we therefore report our results on the thermal conductivity of a few good quality $(Bi_{0.8}Pb_{0.2-y}V_y)_2Sr_2Ca_2Cu_3O_{10+d}$ ($0 = y \leq 0.05$) pellets between 10 and 150 K with the objective of understanding the effect of vanadium substitution in (Bi, Pb) 2223 and the origin of the thermal conductivity maximum below T_c assuming contribution of both electrons and phonons.

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2. Experimental

The usual ceramic route was followed for the synthesis of the present batch of samples, using 99·9% pure Bi_2O_3, PbO, V_2O_5 and CuO and 99% pure $CaCO_3$ and Sr_2CO_3 as starting materials. The above chemicals, in the desired proportions were mixed thoroughly in an agate mortar and then calcined in an alumina crucible for a total period of 48 h at 820 C, with one intermediate grinding after 24 h. The calcined mass was repulverized and then pelletized. The pellets were sintered in air at 840 C for 106 h and subsequently furnace cooled to room temperature.

XRD peaks of the present set of $(Bi_{0·8}Pb_{0·2-y}V_y)_2Sr_2Ca_2Cu_3O_{10+d}$ $(0 = y \leq 0·05)$ samples were indexed according to tetragonal unit cell and they indicate the presence of both 2212 and 2223 phases. But, 2223 phase remains the major one. The lattice parameters obtained corresponding to both 2221 and 2223 phases are given in table 1. The XRD analysis also indicates only marginal changes in the lattice parameters with vanadium addition. Excess oxygen content (d) in the prepared pellets was determined by a volumetric method and all the samples contained nearly the same value of d (table 1).

The electrical resistivity of the samples (12 × 3 × 2 mm) was determined by a d.c. four-probe method. The experiment was performed in an exchange gas cryostat between 77 and 300 K. The temperature was recorded by a calibrated copper-constantan thermocouple. All the voltages were measured with a resolution of 10 nV, using a Keithley model 181 digital nano voltmeter.

A steady state heat flow method, using a pellet shaped specimen (f = 12 mm, thickness = 3 mm), was employed for the measurement of thermal conductivity. The experiment was conducted in a cryorefrigerator (APD model 202) between 10 and 150 K, at a vacuum level better than 10^{-6} torr. One end of the sample was attached to the 10 K stage of the cryo-cooler and the other end to a small copper block containing a 50 Ω constantan wire heater. The pellet was fixed to the upper and lower copper blocks with silver cement adhesive containing high-density fine silver powder. The sample was carefully protected from radiation by a controlled copper radiation shield. Sample temperature was set and controlled by a precision cryogenic temperature controller (Scientific Instrument Model 9650) to a level better than + 0·05 K. A gold + 0·07% iron vs chromel differential thermocouple was used to monitor ΔT. The temperature gradient used in our experiment was between 0·5 and 1 K depending upon the temperature range of measurement. Constant heater power for producing ΔT was provided by a programmable current source (Advantest model TR 6142). All the voltages were measured with a combination of a Keithley model 220 scanner and a digital nano voltmeter (Keithley model 181) with a resolution of 0·01 μV. The maximum estimated uncertainty in the measurement of thermal conductivity was estimated to be 4% over the reported temperature range.

3. Results and discussion

The temperature dependence of the electrical resistivity of the $(Bi_{0·8}Pb_{0·2-y}V_y)_2Sr_2Ca_2Cu_3O_{10+d}$ $(0 = y \leq 0·05)$ pellets between T_{co} and 300 K are given in figure 1. The presence of vanadium in (Bi–Pb) cuprates marginally reduces T_{co} to ~ 104 K (y = 0·05) from 105 K (y = 0). The superconducting onset temperature however remains unaltered (128 K) for all the samples. Resistivity of the samples shows an increase with increasing vanadium content in the sample (table 1). With lowering of temperature, resistivity of all the samples decreases linearly up to close to 130 K with a slope of ~ 0·01 K. Contrary to the XRD data, the temperature dependence of the electrical resistivity of the pellets does not reveal any feature indicative of the 2212 phase.

The thermal conductivity (l) of the above pellets between 10 and 150 K are shown in figure 2. Strong influence of vanadium substitution on the magnitude of l in the measured temperature range is clearly evident. In the normal state at 150 K, the thermal conductivity of the sample with y = 0·05 shows reduction of about 30% with respect to the undoped sample. Similarly, the magnitude of the maximum thermal conductivity (l_{max}) below T_c diminishes to 6·05 mW/cm K for sample with y = 0·05 from 8·85 mW/cm K for sample with y = 0. Other qualitative features of the temperature dependence of the thermal conductivity for both pure and substituted samples are similar to those reported for HTSCs. It is known that the magnitude of the thermal conductivity of HTSCs is primarily dominated by phonon contribution (the electronic contribution being only ~ 20–30%). The increase in elec-

Table 1. Details of the characteristics of $(Bi_{0·8}Pb_{0·2-y}V_y)_2Sr_2Ca_2Cu_3O_{10+d}$ sintered pellets.

Sample composition	T_c (K)	r_{300} (mΩ-cm)	Lattice parameters (Å) 2223 $a = b$	2223 c	2212 $a = b$	2212 c	Excess oxygen (d)
y = 0	105·3	3·25	5·399	36·97	5·360	30·63	0·11
y = 0·025	104·5	3·30	5·403	36·98	5·362	30·63	0·12
y = 0·05	104·0	3·60	5·406	36·97	5·364	30·63	0·12

trical resistivity as well as the small decrease in T_c observed in our vanadium substituted samples suggest that the already small contribution of carriers to the thermal conductivity of (Bi, Pb) 2223 is further reduced due to substitution of vanadium. As the atomic size of vanadium is much smaller than that of Pb, the phonon scattering by mass difference is expected to be enhanced in the present case. In addition, occurrence of sheet-like faults, intergrowths etc are some of the common features in perovskite-like materials and such defects will have frequency dependent phonon scattering rates at lower temperatures. Therefore, the degradation of thermal conductivity observed with vanadium substitution for Pb in (Bi–Pb) 2223, is likely to be associated with the combined effect of decreasing carrier contribution to total l and the overall increase in phonon-defects scattering.

Three different approaches have been proposed till date to account for the occurrence of the thermal conductivity peak below T_c. The phonon approach (Tewordt and Wolkhausen 1989) was the first to be proposed, which was used by many researchers (Dey *et al* 1991; Peacor *et al* 1991; Cohn *et al* 1992; Dey and Barik 1992; Ravindran *et al* 1992; Anderson *et al* 1994; Livanov and Fridman 1994, and many others) to explain the experimental $l(T)$ data with reasonable success. Based on a few experimental evidences (Chawlek *et al* 1990; Nuss *et al* 1991; Bonn *et al* 1992; Romero *et al* 1992), Yu *et al* (1992) argued that the large enhancement in l in the superconducting state originates from the electronic contribution of the Cu–O plane, which increases rapidly below T_c due to strongly suppressed quasiparticle scattering rate in the superconducting state. Assuming l below T_c to be of purely electronic origin, Ausloos and Houssa (1993) also derived an expression for the electronic thermal conductivity (l_e^s) below T_c based on a phenomenological model that includes partial condensation of carriers in HTSCs. It may be noted that various authors (Yu *et al* 1992; Ausloos and Houssa 1993; Houssa and Ausloos 1994; Matsukawa *et al* 1999; Verebelyi *et al* 1999) have been successful in explaining their $l(T)$ data below T_c with the assumption that temperature dependence of the phonon thermal conductivity (l_{ph}) remains unaffected by the superconducting transition. Cohn *et al* (1993) pointed out that such an assumption conflicts with the calculation of l_{ph} for YBCO (Tewordt and Wolkhaussen 1989, 1990; Uher and Ginsberg 1992) using a BCS model for superconductivity. In another approach, Castellazzi *et al* (1997) showed for their sample of BSCCO (2223) *c*-oriented tape that the sudden enhancement in thermal conductivity owes its origin to both phonons and electrons and the thermal conductivity calculated using this approach was in excellent agreement with the experimental data. In view of these, we present here the analysis of our $l(T)$ data taking into account the role of both phonons and electrons towards the origin of the thermal conductivity peak below T_c.

In the temperature range 110–170 K, $l(T)$ is essentially temperature independent. The normal state metallic behaviour of resistivity allows us to estimate the charge carrier contribution to $l(T)$ from the Widemann–Frantz (WF) law:

$$l_e = L_0 T/r,$$

where L_0=2·45 × 10^{-8} WΩK^{-2} is the Lorenz number and r the electrical resistivity. Accordingly, the electronic con-

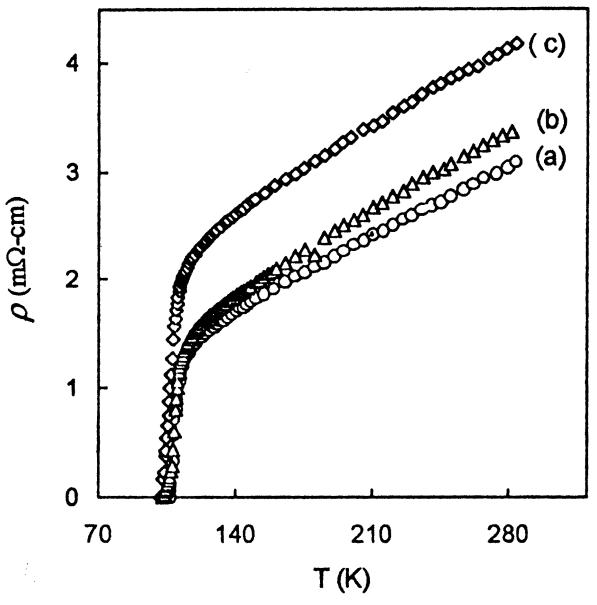

Figure 1. Temperature dependences of the electrical resistivities of $(Bi_{0·8}Pb_{0·2-y}V_y)_2Sr_2Ca_2Cu_3O_{10+d}$ sintered pellets. (a) y = 0, (b) y = 0·025 and (c) y = 0·05.

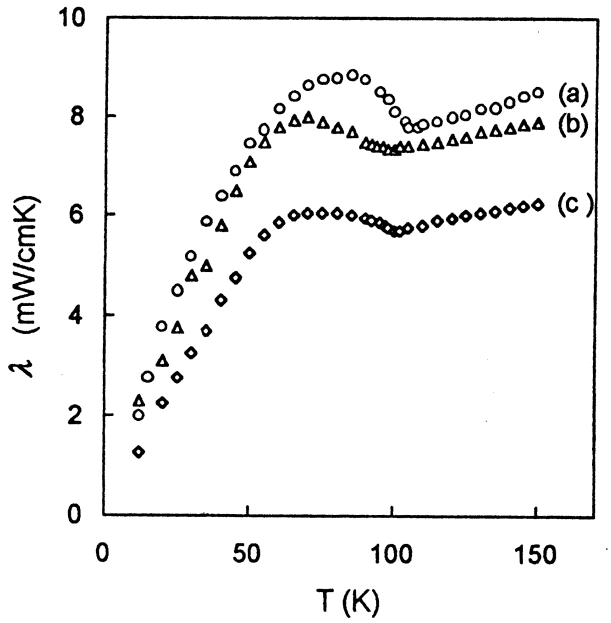

Figure 2. Temperature dependences of the thermal conductivities of $(Bi_{0·8}Pb_{0·2-y}V_y)_2Sr_2Ca_2Cu_3O_{10+d}$ sintered pellets. (a) y = 0, (b) y = 0·025 and (c) y = 0·05.

tribution in the normal state in our samples lies in the range between 24 and 30%. This estimate of l_e may be slightly in error if the electronic relaxation time of the carriers is different for thermal relaxation time, the result of inelastic scattering. However, we take the position as most of the previous researchers have done that the dominant contribution to $l(T)$ above T_c is due to phonon contribution.

The phonon part of the thermal conductivity is calculated using the well-known formulation proposed by Tewordt and Wolkhaussen (1989) which is expressed as:

$$l_{ph} = At^3 \int_0^{q/T} dx \frac{x^4 e^x}{(e^x - 1)^2}$$

$$\times [1 + at^4 x^4 + bt^2 x^2 + dtx + gtxg(x, y)]^{-1}, \quad (1)$$

where, t, x and y refer to the reduced temperature (T/T_c), reduced phonon energy (hw/k_BT) and the reduced energy gap $y = \Delta(T)/k_BT$, respectively. We have taken a scaled BCS gap i.e.

$$\Delta(T) = c\ \Delta_{BCS}(T),$$

where $c = \Delta(0)/\Delta_{BCS}(0)$. Temperature dependence of BCS energy gap is given by

$$\Delta(T) = \Delta(0)[\cos p\ t^2/2]^{1/2}.$$

The coefficient A is related to the boundary scattering and a, b, d and g correspond to the relaxation rates for point defects, sheet like faults, dislocations and electron scattering, respectively. The exact form of the function $g(x, y)$ is given by Bardeen *et al* (1959).

The electronic thermal conductivity (l_e^s) in the superconducting state has been estimated following the expression derived by Yu *et al* (1992) using Kadanoff and Martin (1961) formalism in the clean limit:

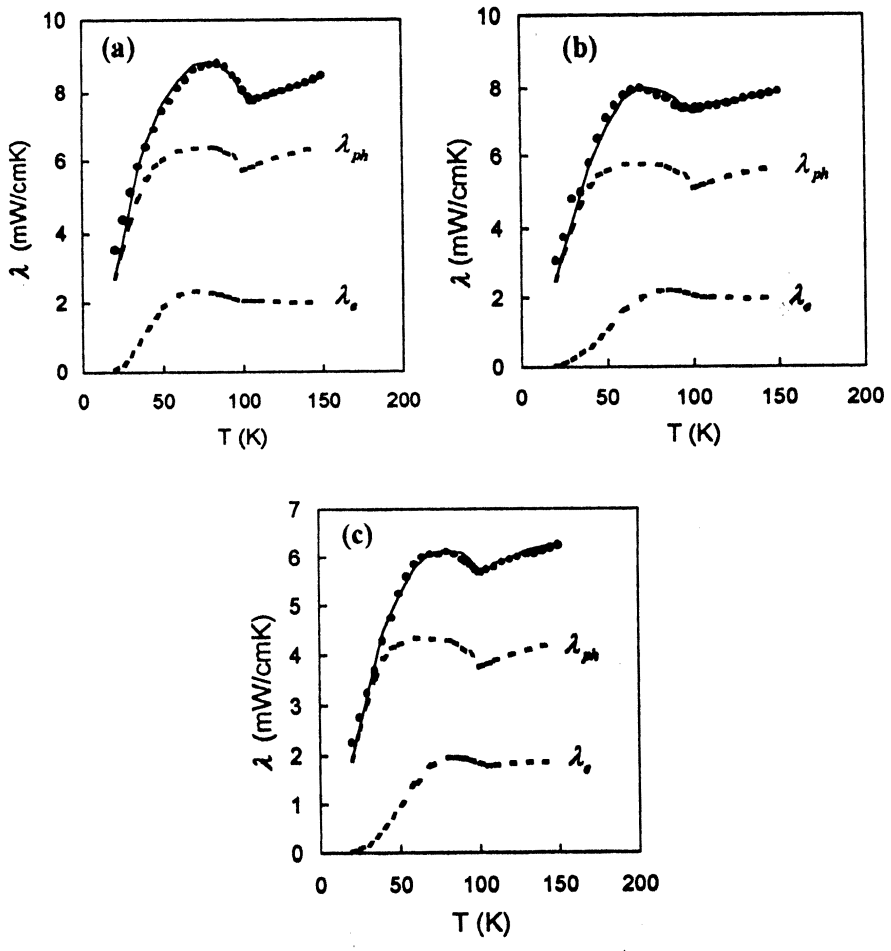

Figure 3. (a)–(c). Points are the experimental data on thermal conductivity of $(Bi_{0.8}Pb_{0.2-y}V_y)_2Sr_2Ca_2Cu_3O_{10+d}$ sintered pellets. (**a**) $y = 0$, (**b**) $y = 0{\cdot}025$ and (**c**) $y = 0{\cdot}05$. l_{ph} and l_e (dashed lines) have been derived from (1) and (2) respectively. Best-fit function (solid line) is the sum of phonon and electron contributions for the parameter values listed in table 2.

$$l_e^s = \frac{N}{2mk_B}\frac{1}{T^2}\int_0^\infty de.e^2.\sec h^2\left(\frac{E}{2k_BT}\right)\frac{1}{\Gamma}, \quad (2)$$

where m is the electron mass, N the electron density and e the normal carrier energy, $E = \sqrt{e^2 + \Delta(T)^2}$ and Ã are the energy and relaxation rate of the quasiparticles respectively. Yu *et al* (1992) defined the reduced quasiparticle relaxation rate without an explicit dependence on energy E as

$$\Gamma'(T) = \Gamma(T)/\Gamma(T_c) \approx t^n + w. \quad (3)$$

The parameter w represents the residual relaxation rate due to impurity scattering. The value of the exponent n depends on the pairing states, viz. $n = 4$ for d-wave pair state.

Thus, in our analysis we take into account the contribution of both electrons and phonons to the thermal conductivity below T_c by using (1)–(3). The adjustable parameters used here are a, b, g, c, A, n and w. $\Delta(0)$ in (1) is adjustable through the parameter c. The results of the best fit for $l(T)$ are shown in figures 3a–c. The best-fit parameters for the present set of samples are given in table 2. The fitted curve is the sum of phonon and electronic thermal conductivity. The separate curves for phonon and electronic contribution to the total thermal conductivity are also shown in figures 3a–c. It may be seen that the calculated values of l give an excellent des-

Table 2. Best fit parameters corresponding to the phonon + electron approach and the estimated values for some of the microscopic quantities for the present set of $(Bi_{0.8}Pb_{0.2-y}V_y)_2Sr_2Ca_2Cu_3O_{10+d}$ superconducting pellets.

Fitted parameter	$y = 0$	$y = 0.025$	$y = 0.05$
A (mW/cmK)	4200	4150	3300
a	1100	1100	1100
b	100	100	100
g	60	80	95
q (K)	300	300	300
$\Delta(0)$ (in k_BT_c units)	1·24	1·24	1·24
n	3	4	4
w	0·10	0·40	0·40
L_b (μm)	20	20	14
l	0·05	0·07	0·11
N_s ($\times 10^5$)	9·08	9·68	13·70
$(\Delta M/M)^2 n$	0·103	0·115	0·165
ΔK ($\times 10^{-4}$ W/cmK)	1·48	4·14	4·42

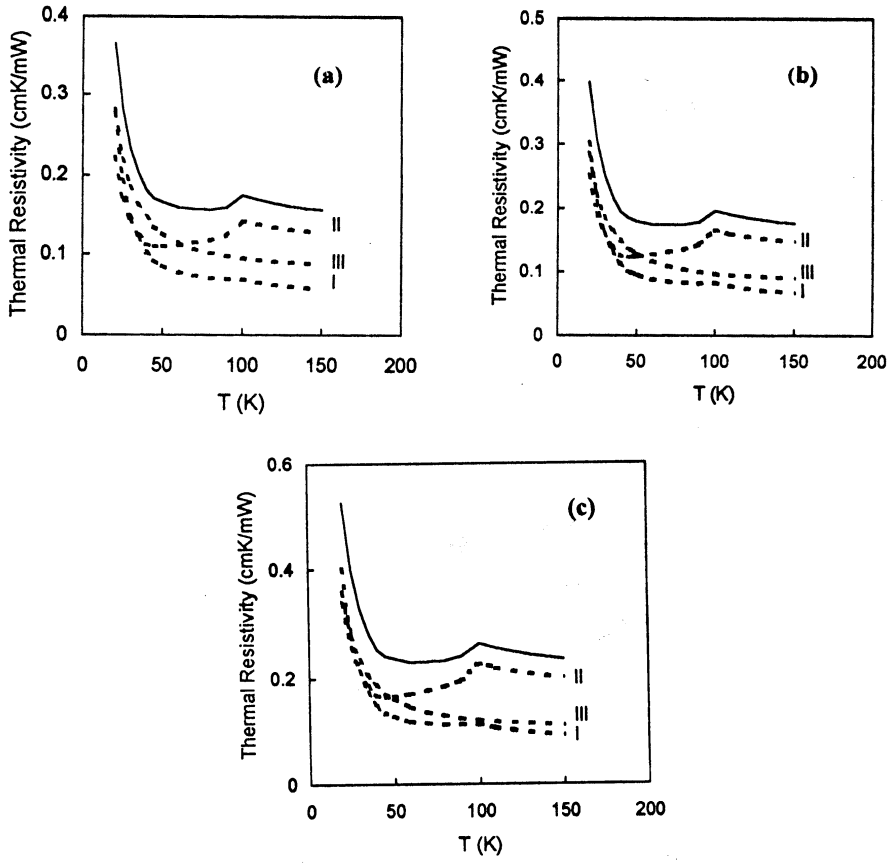

Figure 4. (a)–(c). Temperature dependence of the thermal resistance (solid line) for $(Bi_{0.8}Pb_{0.2-y}V_y)_2Sr_2Ca_2Cu_3O_{10+d}$ sintered pellets. (**a**) $y = 0$, (**b**) $y = 0.025$ and (**c**) $y = 0.05$. Dashed curves are the thermal resistances with the following individual phonon scattering terms removed: (I) point defects, (II) sheet-like faults, and (III) electrons.

cription of the measured data over the entire temperature range from 10–150 K for all the samples. Using the expressions given by Tewordt and Wolkhausen (1989), viz.

$$A = (4p/3)^{1/3}(2p)^{-1}(k_B^2 q/ha^2)(T_c/q)^3 L_b, \quad (4)$$

$$g \cong (p/2)(k_B T_c/\bar{t})(L_b/a)\, l_{la}, \quad (5)$$

where q is the Debye temperature, a an average of the lattice constants, L_b the mean free path for boundary scattering of phonons, t the effective hopping matrix element for a two-dimensional tight-binding band of electrons, and l_{la} the coupling strength between electrons and longitudinal-acoustic phonons. Using the best-fitted values of A and g for our samples, we estimate the values of L_b in the range 10–14 μm and l_{la} between 0·05 and 0·11. In these estimates, we have used q = 300 K, a = 3·8 Å and t = 5000 K. The calculated values of L_b for different samples are within 10–15% of the measured particle size determined from SEM and the value of l_{la} is also comparable with the reported values (Peacor *et al* 1991; Ting *et al* 1994). However, the estimated energy gap parameter Δ(0) is smaller than the BCS value. It may be noted that the fitting procedure as outlined above, is not very sensitive to this parameter and even a larger value of Δ(0) does not have significant influence on the quality of the results reported. It is worthwhile to mention here that the analysis assuming purely electronic contribution to l for $T < T_c$ and taking l_{ph} to remain unaffected by superconducting transition (Yu *et al* 1992; Houssa and Ausloos 1996a), does not yield any reasonable fit to our measured data on pure and vanadium substituted (Bi–Pb)2223 samples.

We further attempted to identify the most dominant scatterers in our samples, particularly near l_{max}. This is shown in figures 4a–c, where we have plotted thermal resistance against temperature with a given scattering process removed, along with the curve for the thermal resistance of the samples corresponding to the best-fit parameters. From such plots we observe that the most dominant scatterers of phonons are the point defects. Using the best-fitted parameters, we further estimated the relative dominance of phonon-defect scattering W_{PD} with respect to all scattering. W_{PD} is defined as

$$W_{PD} = \frac{w_{pd}}{w_{pd} + w_{ps} + w_g},$$

where, $w_{pd} = [1 - l_{ph}/l_{ph}\ (a = 0)]$ and $l_{ph}\ (a = 0)$ are the lattice thermal conductivity calculated from TW equation by omitting the phonon-defect scattering term. It is seen that phonon-defect scattering in the present set of samples constitutes about 50% of the total scattering in the normal state thermal conductivity, which is in agreement with Cohn *et al* (1992) for their YBCO samples.

4. Conclusions

In summary, the thermal conductivity of superconducting $(Bi_{0·8}Pb_{0·2-y}V_y)_2Sr_2Ca_2Cu_3O_{10+d}$ samples has been studied as a function of temperature between 10 and 150 K. Our analysis assuming the role of both phonons and electrons in the formation of peak below T_c gives an excellent agreement with the measured $l(T)$ data for all the samples. Several microscopic quantities calculated from the best-fit parameters also give reasonable values. More sophisticated model that includes a *d*-wave gap parameter and the influence of van Hove singularity on the thermal conductivity (Houssa and Ausloos 1996b; Houssa *et al* 1996, 1998) should help towards better understanding of the role of electrons and phonons in the origin of the observed features of thermal conductivity below T_c in ceramic superconductors.

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