Thermal conductivity of Al-doped MgB₂: Impurity scattering and the validity of the Wiedemann-Franz law

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We report data on the thermal conductivity $\kappa(T,H)$ along the basal plane of the hexagonal crystal structure of superconducting $\mathrm{Mg_{1-y}Al_yB_2}$ with $y\!=\!0.02$ and 0.07 at temperatures between 0.5 and 50 K and in external magnetic fields between 0 and 70 kOe. The substitution of Al for Mg leads to a substantial reduction of the heat transport via electronic quasiparticles. The analysis of the $\kappa(T,H)$ data implies that the Al impurities provoke an enhancement of the intraband scattering rate, almost equal in magnitude for both the σ and the π bands of electronic excitations. This is in contrast with conclusions drawn from analogous data sets for material in which carbon replaces boron and where mainly the intraband scattering rate of the σ band is enhanced. Our complete data set, including additional results of measurements of the low-temperature thermal conductivity of pure MgB₂, confirms the validity of the Wiedemann-Franz law for both pure and doped MgB₂.

DOI: 10.1103/PhysRevB.74.184523 PACS number(s): 74.25.Fy, 74.70.-b

I. INTRODUCTION

In multiband superconductors, the onset of superconductivity may introduce gaps of different size on different parts of the Fermi surface. As a consequence, the thermal conductivity of multiband superconductors at $T < T_c$, the critical temperature, and in external magnetic fields $H \leq H_{c2}$, the upper critical field, reveals specific features that are usually not observed for common type-II superconductors. These features were first reported for MgB₂ (Ref. 1) and subsequently also identified for NbSe₂ (Ref. 2) and PrOs₄Sb₁₂ (Ref. 3). The thermal conductivity $\kappa(H)$ of these materials in the mixed state of superconductivity, measured at constant temperature, exhibits an unusual two-step increase with increasing magnetic field between H_{c1} and H_{c2} . The two steps reflect the separate recovery of the heat transport carried by quasiparticles of two electronic energy bands with significantly different energy gaps in the superconducting state.^{4,5} In order to observe this type of behavior of $\kappa_{e}(H)$, the electronic contribution to the total thermal conductivity, it is important that in the corresponding temperature and magneticfield ranges, the heat transport carried by phonons and represented by $\kappa_{\rm ph}(H)$ is relatively weak. In addition, also the conditions of weak interband scattering of the quasiparticles and a scattering of quasiparticles in the band developing the smaller gap compatible with a moderately dirty limit situation need to be fulfilled.⁴ All these conditions are ideally met for MgB2 at helium temperatures and the two step enhancement of $\kappa_e(H)$ with increasing field is very pronounced, such that the contribution of each of the two types of bands to κ_e can be identified. Hence this allows us, in principle, to investigate the influence of different perturbations on the heat transport carried by different energy carriers separately. Indeed, results of measurements of the thermal conductivity of $Mg(B_{1-r}C_r)_2$ (Ref. 6) revealed that low concentrations of C replacing B reduce the steplike enhancement of $\kappa(H)$ near H_{c2} much more than the analogous feature of $\kappa(H)$ which is observed at much lower fields. This result suggests that the carbon related defects scatter quasiparticles of the band with the large gap much more than quasiparticles excited in the band with the smaller gap. This observation agrees with the expectation that C substitutions for B mostly enhance the scattering between states in the σ band which is predominantly formed by boron sp_xp_y orbitals. In this work we describe and discuss results of analogous measurements of $\kappa(H,T)$ of $\mathrm{Mg}_{1-y}\mathrm{Al}_y\mathrm{B}_2$. The investigations of single-crystalline material with y=0.02 and 0.07 reveal that Al replacing Mg at the few at. % level reduces the heat transport of quasiparticles in both the σ and the π bands in a more or less equal manner.

Aluminium replacing Mg in MgB₂ is expected to influence the superconductivity of the pure binary compound mainly via two effects.^{7,8} First, Al doping causes a reduction of hole-type carriers, thus reducing the corresponding density of electronic states (DOS) and, likewise, the size of both gaps Δ_{σ} and Δ_{π} , thereby accounting for the reduction of T_c . The second consequence is caused by out-of-plane distortions created by Al occupying Mg sites which enhance the interband scattering thus leading to a relative increase of the gap in the π band. Indeed, results of various experimental studies confirm this view and suggest that for $y \leq 0.1$, the replacement of Mg by Al leads to a gradual reduction of Δ_{σ} without substantial changes in the value of Δ_{π} . For y >0.1 the situation is more controversial but most experimental results are compatible with the reduction of both gaps upon increasing $y.^{10,12,13}$ Because choosing y>0.1 often leads to inhomogeneities in the Al distribution, such as the segregation of samples into Al-rich and Al-poor regions, 12,14 we restricted our study to material with y < 0.1, for which the influence of Al is expected to be as described above.

II. EXPERIMENT

The preparation and characterization of the $Mg_{1-y}Al_yB_2$ single crystals that were used in this study is described in detail in Ref. 14. The synthesis under high pressure allowed the production of single-phase, single-crystalline material up to Al concentrations $y \sim 0.1$. For our measurements we selected two samples, below denoted as Al2 and Al7, with Al contents, as determined by energy-dispersive x-ray analysis,

TABLE I. Samples of $Mg_{1-y}Al_yB_2$ studied in this work. Sample MB0, listed for comparison, was studied earlier (see Ref. 1).

	у	T_c (K) from $\rho(T)$	$\rho(40 \text{ K}) \\ (\mu\Omega \text{ cm})$	ρ(300 K)/ρ(40 K)
MB0	0	38.1 ± 0.1	2.05	6.80
MB1	0	38.3 ± 0.1	0.71	7.49
MB2	0	38.2 ± 0.1	1.20	7.11
A12	0.017	35.9 ± 0.15	2.36	3.90
Al7	0.073	33.0 ± 0.2	4.13	2.70

of y=0.02 and 0.07, respectively. Since we also intended to confirm or refute our earlier conclusion of the nonvalidity of the Wiedemann-Franz law for MgB₂ at helium temperatures (Ref. 1), we repeated the measurement of the thermal conductivity of binary MgB2, using two newly synthesized single crystals, denoted as MB1 and MB2 below. All samples were bars with the approximate shape of a prism and typical dimensions of $0.9 \times 0.3 \times 0.06$ mm³. The thermal conductivity was measured using the same standard uniaxial heat-flow method that we applied in our previous experiments on pure and C-doped MgB₂. In this setup, one end of the sample is attached to a copper body serving as a heat sink and at the opposite end, a miniature RuO2 resistor, serving as the heater, is attached. The temperature gradient along the sample was monitored employing either RuO₂ resistor thermometers below 4 K or chromel/Au+0.07% Fe thermocouples above 4 K. These temperature sensors were each attached to a 25-\(\mu\)m-diameter gold wire and the wires were glued to the crystals with silver-filled epoxy, approximately 300 to 400 μ m apart along the direction of the heat flow. The temperature difference along the sample was chosen to be of the order of 3% of the average sample temperature. For the subsequent measurements of the dc electrical resistivity ρ , a four contact configuration was used. In order to minimize the uncertainty of the geometrical factor in the comparison of the electrical and thermal conductivities, the described thermometer contacts were also used as voltage contacts for the resistivity measurements. Nevertheless, the possible systematic errors in the separate evaluation of κ or ρ values caused by the uncertainty of the sample geometry were as large as $\pm 20\%$, whereas the random errors for κ and ρ were only $\pm 1\%$ and $\pm 0.1\%$, respectively.

III. RESULTS

The electrical resistivity was measured between 2 and 300 K in zero magnetic field and H=50 kOe, oriented along the crystallographic c direction. For both new specimens of pure MgB₂, MB1, and MB2, $\rho(T)$ is very similar to the data that we obtained in earlier work (Ref. 1) for sample MB0. Likewise the values of T_c , the transition widths, and the residual resistance ratios turned out to adopt similar values as were established in our previous work. Note, however, that the absolute resistivities of the new samples just above T_c are somewhat lower than before (see Table I). In the temperature range between T_c and approximately 150 K, $\rho(T)$ may very

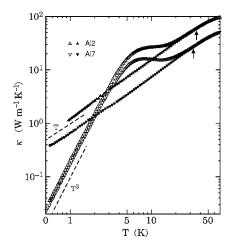


FIG. 1. Thermal conductivity vs temperature in the ab plane of single-crystalline $\mathrm{Mg}_{1-y}\mathrm{Al}_y\mathrm{B}_2$ (y=0.02 and 0.07) in zero magnetic field (open symbols) and $H\parallel c$ =50 kOe (solid symbols). The arrows indicate the critical temperatures in zero magnetic field.

well be represented by $\rho(T) = \rho_0 + aT^3$. The most obvious variation of ρ upon replacing small amounts of Mg by Al is an expected increase of the residual resistivity ρ_0 , already reached at 40 K, which is caused by an enhanced scattering rate due to the Al defects in the Mg sublattice. The temperature-dependent component due to the scattering of electrons by phonons changes only very little.

In Fig. 1 we display the thermal conductivity of $\mathrm{Mg}_{1-y}\mathrm{Al}_y\mathrm{B}_2$ (y=0.02 and 0.07) as a function of temperature in zero magnetic field and for H=50 kOe, with H oriented parallel to the c axis. In zero magnetic field, the absolute difference in magnitude of κ is more pronounced at elevated temperatures but with decreasing temperature to much below T_c this difference is reduced significantly and is only marginal at temperatures below 3 K. Below 3 K, the temperature dependence of κ is fairly well represented by $\kappa(T) \propto T^3$. In the field induced normal state, the relative difference in the κ values persists to temperatures below 1 K where $\kappa(T)$ approaches a variation linear in temperature.

In Fig. 2 we display typical curves for $\kappa(H)$, obtained for the two investigated samples at various constant temperatures. Below H_{c1} and above H_{c2} , the thermal conductivity varies only weakly with field. However, in the mixed state, i.e., between H_{c1} and H_{c2} , $\kappa(H)$ exhibits a substantial field dependence, as expected. While H_{c1} cannot reliably be determined from our data sets because the $\kappa(H)$ curves are irreversible in the vicinity of H_{c1} , the upper critical field can be identified quite accurately from the abrupt change of slope of $\kappa(H)$, as is indicated by the arrow in the upper left panel of Fig. 3. The resulting curves of $H_{c2}(T)$ are shown in Fig. 3. We emphasize, as in our previous work, that $H_{c2}(T)$ obtained in this manner is a reliable measure for the temperature dependence of the upper critical field of the bulk of the sample. insensitive to surface superconductivity or minority-phase inclusions. 15 The observed trend of the alteration of $H_{c2}(T)$, its values, and anisotropies (see inset of Fig. 3) upon Al concentration, is consistent with the results presented in Ref.

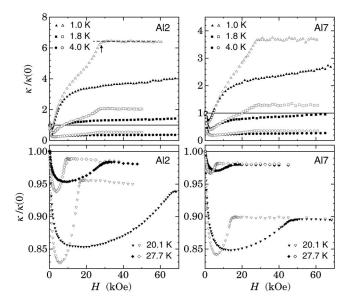


FIG. 2. Thermal conductivity in the basal plane of $Mg_{1-y}Al_yB_2$ (y=0.02,0.07) vs H at several fixed temperatures. The closed and open symbols correspond to the field directions perpendicular and parallel to the c axis, respectively. The arrow in the upper left panel demonstrates how H_{c2} for a given field orientation and at a given temperature was established.

IV. ANALYSIS AND DISCUSSION

A. Multiband feature of thermal transport

In MgB $_2$ the total heat transport is carried by itinerant lattice and electronic quasiparticle excitations and κ is given by the sum

$$\kappa_{\text{tot}} = \kappa_{\text{ph}} + \kappa_e$$
 (1)

with $\kappa_e = \kappa_{e,\sigma} + \kappa_{e,\pi}$, where the last two terms capture the heat transport by quasiparticles of the σ and the π bands, respectively. In principle, fitting the zero-field $\kappa(T)$ data in the temperature range $T \le T_c$ by invoking Eq. (1) and the appropriate theoretical expressions for the individual terms, estimates of the corresponding gaps in the quasiparticle excitation spectra can be made. The theory of Bardeen, Rickayzen, and

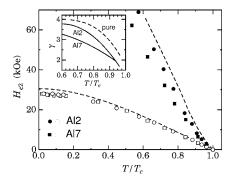


FIG. 3. The upper critical fields $H_{c2}(T/T_c)$. The closed and open symbols correspond to the field directions perpendicular and parallel to the c axis, respectively. For comparison, our earlier results for pure MgB₂ (Ref. 15) are shown by the dashed lines. The inset shows the anisotropies of the upper critical fields $\gamma = H_{c2}^a/H_{c2}^c$.

Tewordt (Ref. 16) has successfully been used to evaluate the energy gaps of conventional, single-gap superconductors from thermal conductivity data. For multigap superconductors, more than one channel of quasiparticle heat transport in different energy bands need to be considered¹⁷ and therefore the number of fit parameters increases according to the increasing number of bands involved. If the heat transport carried by phonons is not negligible, the number of independent parameters increases even more. In particular, also the scattering of phonons by quasiparticles has to be taken into account. In our case, the upturn of $\kappa(T)$ in zero magnetic field just below T_c indicates that the phonon contribution to the thermal conductivity in the normal state, κ_{ph}^n , is not small in comparison with the normal-state component κ_e^n . Thus $\kappa_{\rm ph}$ is not negligible at $T \lesssim T_c$ and any fitting procedure in the manner indicated above seems unreasonable. Nevertheless, in Ref. 18 an attempt of such an analysis has been made for data obtained for polycrystalline MgB₂ where the phonon contribution to the total thermal conductivity is claimed to be negligibly small.

Information about details of quasiparticle scattering can be obtained from the analysis of the magnetic-field dependence $\kappa(H)$ of the thermal conductivity, measured in the mixed state of type-II superconductors at constant temperature. Usually, upon increasing H above H_{c1} , $\kappa(H)$ first decreases because of an additional scattering of both phonons and quasiparticles by vortices. With still increasing H, the increasing density of quasiparticles in normal vortex cores enhances κ_e . Also for MgB₂ the thermal conductivity in the mixed state is dictated by such processes but the resulting features of $\kappa(H)$ are clearly different from those observed for common superconductors. First, κ_e increases very rapidly in rather low fields, such that a considerable part of the normalstate thermal conductivity is recovered already when H $\ll H_{c2}$. This is distinctly different from observations in common type-II superconductors where, depending on the sample purity, $\kappa_e(H)$ either grows significantly and rapidly only if H approaches H_{c2} (pure limit) or reaches its normalstate value in an approximately linear fashion, i.e., κ_e $\propto H/H_{c2}$ (dirty limit). ¹⁹ Second, when measured along different crystallographic directions and taking into account the anisotropy of the upper critical field, the low-field increase of $\kappa(H)$ does not scale as H/H_{c2} as is the case for common type-II superconductors. For MgB₂, these anomalous features find a natural explanation by considering the anisotropy of the gap in the quasiparticle excitation spectrum, i.e., the different gap values for the σ and the π bands, respectively. It turns out that the smaller gap Δ_{π} is quenched already at $H^* < H_{c2}$, where the critical field H^* is practically independent of the orientation of H, reflecting the three-dimensional nature of the π band. In this scenario, in the intermediate field regime $H \approx H^* < H_{c2}$, the contribution of the quasiparticles created in the π band is close to that in the normal state whereas in the σ band, the large gap prevents the excitation of quasiparticles until H approaches H_{c2} and hence $\kappa_{e,\sigma}$ grows substantially only when H is close to H_{c2} . In shape this is similar to what is observed for common, moderately pure type-II superconductors. Considering that at a given temperature, H_{c2}^{ab} is significantly larger than H_{c2}^{c} , the ratio

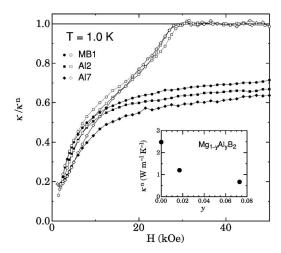


FIG. 4. Reversible thermal conductivity κ normalized to its normal-state value κ^n vs H at 1.0 K for pure and Al-doped MgB₂. The closed and open symbols correspond to the field directions perpendicular and parallel to the c axis, respectively. The inset shows doping dependence of κ^n at 1.0 K.

 $\kappa_e(H \perp c \approx H^*)/\kappa_e(H \parallel c \geq H_{c2})$ provides a rough estimate of the ratio $\kappa_{e,\pi}^n/\kappa_e^n$.

In Fig. 4 we compare $\kappa(H)$, measured at T=1.0 K, for pure and Al-doped MgB₂; only data points where $\kappa(H)$ is reversible are included in the plot. In this temperature range, $\kappa_{\rm ph} \ll \kappa_e^n$ and therefore the field dependence of κ mostly reflects that of the thermal conductivity due to quasiparticles. The diagram clearly indicates that Al substitutions for Mg reduce the ratio $\kappa_{e,\pi}^n/\kappa_e^n$ and thus $\kappa_{e,\pi}^n/\kappa_{e,\sigma}^n$ but the latter ratio changes much less than the total electronic thermal conductivity in the normal state κ_e^n (see inset of Fig. 4). For the ratios $\kappa_{e,\pi}^n/\kappa_{e,\sigma}^n$ at 1 K, we estimate the values of 1.9, 1.6, and 1.2 for the samples MB1, Al2, and Al7, respectively. This suggests that although the introduction of Al for Mg reduces the thermal conductivity of quasiparticles slightly more strongly in the π band, the overall effect is nearly the same in both channels. This is in stark contrast to the observations made in the case of $Mg(B_{1-x}C_x)_2$ where a 3% substitution of B by C reduces the quasiparticle thermal conduction of the σ band to a negligible level in comparison to the π band contribution.

The general equations capturing the thermal conductivity due to quasiparticles can be simplified to the form

$$\kappa_e = \frac{1}{3} C_{Ve} v_F l_e, \tag{2}$$

where C_{Ve} is the electronic specific heat, v_F is the Fermi velocity, and l_e is the electronic mean free path. In the mixed state the mean free path is given by $l_e = v_F/(\gamma + \gamma_A)$, where γ is the scattering rate due to impurities and γ_A is the scattering rate due to Andreev scattering of quasiparticles at vortices. ¹⁷ In this scenario, only γ is affected by Al impurities. For MgB₂ this approach has to be modified to

$$\kappa_{e,i} = \frac{1}{3} C_{Ve,i} v_{F,i} l_{e,i},$$
(3)

with $i=\pi$, σ . It has been established that Al substitutions up to y=0.1 change neither C_{Ve} nor $v_{F,\pi}$ in the ab plane in any

significant way.¹³ With an Al content y=0.1, $v_{F,\sigma}$ in the ab plane is reduced only by 10-15 % with respect to its value in the pure compound.^{10,13} This suggests that the reduction of the quasiparticle mean free path l_e is responsible for the substantial reduction of κ_e shown in the inset of Fig. 4. If we assume that both the specific heat and the Fermi velocities remain indeed unchanged, our data imply that a 7.3 at. % substitution of Mg by Al leads to reductions of $l_{e,\sigma}$ and $l_{e,\pi}$ by factors of 2.7 and 4.5, respectively. A de Haas-van Alphen study of Mg_{1-y}Al_yB₂ (Ref. 20) indicates that for $y \approx 0.075$, the mean free path on the σ sheet of the Fermi surface is approximately a factor of 2 shorter than for pure MgB₂ and for the π sheet, the reduction of $l_{e,\pi}$ is estimated to be of the order of 3 or slightly larger. These results are obviously in very good agreement with our conclusions.

B. Relations between thermal and electrical conductivity

The relaxation rates which govern the electronic charge and heat currents in metals, respectively, are in general not identical. However, some of the involved scattering processes are, under certain conditions, equally efficient in relaxing both electric and thermal currents. In these cases, the electrical resistivity ρ and the electronic thermal conductivity κ_e are related via the law of Wiedemann and Franz (WFL), such that

$$\kappa_{\rho}(T) = L_0 T / \rho(T), \tag{4}$$

where L_0 =2.45 × 10⁻⁸ W Ω K⁻² is the Lorenz number. This relation is expected to be valid for any metal whose electronic subsystem can be described as a Fermi liquid. Elastic scattering of electrons by static lattice defects is one of the mechanisms which are compatible with the validity of the WFL. If this type of scattering dominates all other processes, the residual resistivity at low temperatures adopts a constant value and, in the same temperature regime, κ_e varies linearly with temperature. The experimental Lorenz number $L(T) = \kappa(T)\rho(T)/T$, with $\kappa(T)$ as the directly measured thermal conductivity, is a constant and equal to L_0 only if the phonon thermal conductivity is negligible, i.e., $\kappa_{\rm ph} \ll \kappa_e$ and hence $\kappa_e \approx \kappa$. Inelastic-scattering processes always lead to $L(T) < L_0$ and sizable heat transport by phonons provides and excess heat conduction and hence $L(T) > L_0$.

In the field induced normal state, all samples of MgB₂ exhibit a temperature-independent resistivity below 40 K, suggesting that defect scattering of quasiparticles dominates the low-temperature electronic transport. In passing we note that even in external magnetic fields, oriented along the c axis and of the order of 50 kOe, i.e., exceeding the corresponding bulk value of H_{c2} at T=0 K for all investigated materials, the onset of a superconducting transition is observed at very low temperatures. Most likely this transition is related to properties of the sample surface and hardly affects the predominantly probed heat transport in the bulk of the sample. In order to check the validity of the WFL, we thus used ρ = ρ_0 \approx ρ (40 K), presented in Table I.

In part of our earlier work¹ we encountered evidence that for pure MgB₂, $L(T)/L_0 > 1$ at low temperatures which could

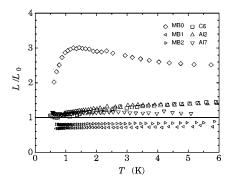


FIG. 5. Normalized Lorenz number $L(T)/L_0$ for three samples of pure MgB₂ and for C- and Al-doped MgB₂. C6 denotes Mg(B_{0.94}C_{0.06})₂ (data from Ref. 6).

not simply be attributed to a substantial phonon contribution to $\kappa(T)$, at least not below T=6 K. Our most recent experiments on $\mathrm{Mg}(\mathrm{B}_{1-x}\mathrm{C}_x)_2$ single crystals, however, gave no evidence for a violation of the WFL in the same temperature region. In Fig. 5 we plot these earlier results together with the corresponding new data that we accumulated for Aldoped MgB₂. Below 1 K, the series of both C and Al doped materials exhibit ratios L/L_0 very close to 1. The smoothly increasing deviation from the ideal value with increasing temperature can easily be attributed to a non-negligible κ_{ph} which, in the normal state of a metal exhibits a temperature variation that is typically stronger than linear in T.

Since it appeared that the WFL is well satisfied in MgB_2 with small amounts of defects due to Mg and B substitutions in the form of Al and C, respectively, it seemed of interest to verify whether the reported violation of the WFL is limited to pure MgB_2 . We thus reexamined the electrical and the thermal transport properties of two new single crystals of MgB_2 (MB1 and MB2) between 0.5 and 6 K, both in the superconducting state in zero magnetic field and in the normal state, induced by a magnetic field of 50 kOe, oriented along the c direction of the crystal lattice. The corresponding data resulted in the L/L_0 ratios that are also displayed in Fig. 5.

In contrast to the substantial and temperature-dependent enhancement of the Lorenz number that resulted from the data that were earlier obtained for sample MB0, we now note an almost temperature-independent L(T), somewhat smaller than L_0 , for both new samples of pure MgB₂. The reduction of L(T) to below the value of L_0 is most likely not caused by inelastic scattering of the quasiparticles, because an almost parallel shift of L(T) from L_0 would require that the responsible scattering would add a temperature-independent contribution to the thermal resistivity. However, all inelastic-scattering processes that we are aware of, vary their strength with temperature. A more plausible explanation is to attribute

the constant shift of L(T) from L_0 to uncertainties in the sample geometry. If, as in our experiments, the same contacts are used to measure either the voltage or the temperature difference along the sample in the respective measurements of the electrical or the thermal conductivity, the same geometrical factor applies for the corresponding equations for calculating ρ or κ . Thus in calculating L(T), this factor drops out. This is really only true for cases where the two contacts can be mounted far apart, in the sense that their separation d is much larger than the contact width δ . Our samples are quite small and hence $d \approx \delta$. In this case it is quite possible that the effective distances between the contacts for measuring the voltage or the temperature difference, i.e., d_0 or d_{κ} , are different. If now the ratio d_{κ}/d_{ρ} differs from 1, the calculation of L will result in a number that differs from L_0 . For our small crystals, the distance between the contacts was of the order of 300-400 μ m and the contact widths varied between 50 and 70 μ m. It is therefore not unlikely that d_{κ} $\neq d_0$ which, in turn, may explain the constant shift of L(T)from L_0 .

In this sense we believe that no anomalous violation of the WFL is observed for the two new samples of single crystalline MgB_2 . This suggests that our earlier observation reported in Ref. 1 is not an intrinsic property of the material. We note, however, that the $\kappa(H)$ data exhibit the same features for all the investigated samples of pure MgB_2 and therefore all the conclusions concerning the two-band features of MgB_2 drawn in Ref. 1 remain valid.

V. CONCLUSIONS

Based on experimental results of the electrical and thermal conductivity we note that the substitution of magnesium by aluminium in MgB₂ provokes an increase of the quasiparticle scattering of similar magnitude in both the σ and the π bands of electronic states. The calculated reductions of the quasiparticle mean free paths in the σ and π bands, respectively, are in agreement with results of de Haas–van Alphen experiments. The considerable violation of the Wiedemann-Franz law at low temperatures for pure MgB₂ that we reported in Ref. 1 could not be confirmed in additional experiments using new single crystals of this compound. Since also for all the doped MgB₂ crystals that we studied since (Ref. 6 and this work), no anomalous deviations of the WFL were identified, our previous observation must have been of extrinsic origin.

ACKNOWLEDGMENT

This work was financially supported in part by the Schweizerische Nationalfonds zur Förderung der Wissenschaftlichen Forschung.

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