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Heat conductivity of amorphous PdCuSi alloys containing Fe and Mn impurities

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In a search for the existence of magnetic contributions to the phonon thermal resistivity, the low-temperature heat conductivity $\kappa(T)$ (0.3 – 10 K) of an amorphous diamagnetic $Pd_{77.5}Cu_6Si_{16.5}$ sample has been compared with $\kappa(T)$ of amorphous spin-glass-like $(Pd_{77.5-x}Cu_6Si_{16.5})$ M_x alloys, containing $3d$ transition metals with $x = 3$ at. % Mn and $x = 1$ at. % Fe. The phonon thermal conductivities $\kappa^{ph}(T)$ of the magnetic alloys show roughly the same temperature dependence as the PdCuSi matrix. However, κ^{ph} in these alloys is between 20% and 30% smaller as compared with PdCuSi. This can be explained within the tunneling-state model by an enhanced density of two-level systems (TLS) in the magnetic samples. Influences on κ^{ph} due to either spin freezing in the range around T_f or to external magnetic fields up to 5 T could not be detected. Therefore evidence for the existence of extrinsic magnetic TLS cannot be inferred from the present experiments.

I. INTRODUCTION

To date, there is general belief that the low-temperature physical properties of amorphous materials are influenced by low-energy excitations associated with the disordered structure. A simple physical description for these excitations is given within the so-called two-level – system model (TLS), developed independently by Anderson *et al.*[1] and Phillips.[2] In particular, one assumes that certain atoms or groups of atoms reorient via quantum-mechanical tunneling between two configurationally determined energy levels. The resulting localized excitations provide an excess specific heat, increasing linearly with temperature below ~ 1 K. They also act as phonon scatterers, giving rise to a typical roughly T^2 dependence of the phonon thermal conductivity also in the temperature range below 1 K. Both features are experimentally well established in a variety of nonmetallic and metallic amorphous materials.[3–7] However, despite the generality of these experimental observations, a satisfactory understanding of the atomistic nature of the TLS is still lacking.

There have been many attempts to prove whether the concentration of the TLS depends on the presence of certain types of impurities or defects. In this context it has been shown that, e.g., the addition of OH^- to vitreous silica led to an enhancement of the low-temperature specific heat,[8] while the thermal conductivity remained almost unchanged. An explanation for this has been recently given by Golding *et al.*[9] on the basis of dielectric echo experiments on fused silica doped with different amounts of OH^-. It was shown that one can distinguish between *intrinsic* TLS, with small electric dipolar moments and strong coupling to phonons, and *extrinsic* TLS, associated with the OH^- impurities, having large electric dipolar moments, but much weaker scattering strength for phonons. Both types of TLS contribute to the specific heat, but mainly intrinsic TLS contribute to the thermal conductivity.

Experiments to distinguish between intrinsic and extrinsic TLS by adding *magnetic* impurities to dielectric glasses failed. In, e.g., manganese aluminosilicate glass the low-temperature specific heat is dominated by the contribution from magnetic excitations[10] so that the small contribution from the TLS cannot be separated. On the other hand, measurements of the low-temperature thermal conductivity on the same glasses[11] showed no extra contribution from the Mn impurities. Moreover, efforts to alter the thermal conductivity by applying a magnetic field of about 5 T on a vitreous silica sample containing 0.5 wt. % Fe_2O_3 also failed.[12]

Our investigation concerns the question whether in addition to intrinsic TLS possibly extrinsic TLS exist in *metallic* glasses. We have done this by ad-

ding magnetic impurities to the widely studied PdCuSi amorphous alloy. This matrix material was chosen because it is diamagnetic and can easily be transformed into the glassy state (low quenching rate), allowing rod-shaped samples with well-defined geometry and consequently small errors in the geometry factor for our measurements of the heat conductivity. Furthermore, Pd can be replaced by $3d$ magnetic impurities to relatively high concentrations without disturbing the glass formation. From our own low-temperature susceptibility studies[13] and from earlier experiments,[14] it is known that PdCuSi containing several atomic percent of Fe, Mn, or Co shows spin-glass-like transitions. We can thus also study the question whether or not this magnetic transition is detectable in the thermal transport.

The experiments will show that blocking of spins around T_f does not affect the heat conductivity in PdCuSi alloys doped with Mn and Fe. There is also no influence of a magnetic field (up to 5 T) to be seen in $\kappa(T)$. We observe, however, an enhanced phonon thermal resistivity in the magnetic samples with respect to the nonmagnetic matrix.

II. EXPERIMENTAL

The measurements have been carried out between 0.3 and 10 K in an all-metal, single-shot ^{3}He cryostat, with a 5-T superconducting magnet immersed in the 4-K bath. The heat conductivity is measured on sample rods of well-defined geometry (diameter 1 mm, length 4 cm) in the following way. The upper end of the sample is clamped onto the ^{3}He bath, whose vapor pressure is electronically stabilized by an electromagnetic bypass valve and a resistance heater. Although the cryostat is only single shot, the setup allows measurements over extended periods of time with long term temperature stability of $\delta T/T < 10^{-4}$. To achieve a temperature gradient along the sample, a resistance heater, attached to its lower end, produces a constant heat current $\dot{Q}$ through the sample cross-section area A. The resulting ΔT is measured over a distance l by two thermometers thermally anchored to the sample by small gold-plated Cu clamps. The total conductivity $\kappa^{\text{tot}}(T)$ is determined by

$$\kappa^{\text{tot}}(T) = \frac{l}{A}\frac{\dot{Q}}{\Delta T - \Delta T_0} , \tag{1}$$

where ΔT_0 is the temperature difference measured for zero heater power.

For thermometry below 1 K, we use two carbon chips, cut from Speer 220-Ω $\frac{1}{2}$-W resistors, and above 1 K, two carbon-glass resistors (CGR 250). All thermometers are calibrated against Lake Shore Cryogenics Ge resistors. Calibration in the magnetic field is achieved by keeping the Nb-shielded Ge resistors out of the field. Carbon and carbon-glass resistors are used because of their isotropic and small magnetoresistance as compared to, e.g., Ge resistors. Accuracy and reproducibility of the temperature measurements are of the order of 10^{-3}. Calibration check of the thermometry is done after every third run.

Since the heat conductivity of the amorphous metal samples is very low one has to be sure not to thermally shunt the samples by the electrical leads. Therefore, we use single-filament NbTi superconducting wire of 50-μm diameter. This keeps the error in the measured thermal conductivity due to shunting below 1%, whereas the error due to shunting by the residual gas is smaller than 0.1% for a typical vacuum of 6×10^{-7} mbar. The temperature is determined by conductivity bridges limiting the heat influx into the sensing elements to a maximum of 10^{-10} W, i.e., $\sim$0.5% of the lowest total heat input into the samples.

In conclusion, the relative uncertainty in the heat conductivity measurements from sample to sample can be kept below $\pm$7%. However, κ may contain a systematic error up to 20%, because the effective distance of the temperature-sensing elements may differ from the distance l [Eq. (1)] due to the final width of the copper clamps.

Alloys are prepared from palladium, copper, and silicon, all of 99.999% purity (supplied by Goodfellow Metals), and 99.99% pure iron and manganese. The components are melted in an induction furnace and then put into a quartz tube and degassed and homogenized for 6 h at about 1000 K. The alloys are then remelted and pressed with argon gas into the thin-walled lower capillary end (1-mm i.d.) of the melting quartz tube and rapidly quenched in water. The sample structure is checked by x-ray diffraction with Cu $K\alpha$ radiation. Besides a broad peak, typical for the amorphous short-range order, no Bragg peaks are detected. A further check for the amorphous state is the measurement of the residual resistivity ratio (RRR) ρ(300 K)/ρ(4.2 K). Values of the RRR and of the absolute resistivity at room temperature are given in Table I. They are consistent with what has been found by other groups.[5,15] The magnetic state of the samples con-

TABLE I. Characteristic data for the investigated samples. Density; electrical resistivity at room temperature ρ(300 K); residual resistivity ratio (RRR) ρ(300 K)/ρ(4.2 K); spin-glass-freezing temperature T_f (K); coefficients α and β as taken from a fit: $\kappa^{ph} = \alpha T^{\beta}$ for temperatures below 1 K. The last column gives the ratio of κ^{ph}(1 K)/κ^{tot}(1 K).

	Density (g/cm^3)±0.05	Resistivity at 300 K ρ ($\mu\Omega$ cm)	Residual resistivity ratio	Freezing temperature T_f (K)
$Pd_{77.5}Cu_6Si_{16.5}$	10.54	66.78	1.036	
$Pd_{76.5}Cu_6Si_{16.5}Fe_1$	10.61	61.57	1.210	2.15
$Pd_{74.5}Cu_6Si_{16.5}Mn_3$	10.50	100.46	1.036	0.85

taining Mn and Fe is characterized by ac susceptibility measurements, showing spin-glass-like behavior with cusp temperatures T_f as listed in Table I.

III. RESULTS AND DISCUSSION

Figure 1 shows the total heat conductivity κ^{tot} (open circles) versus temperature in logarithmic

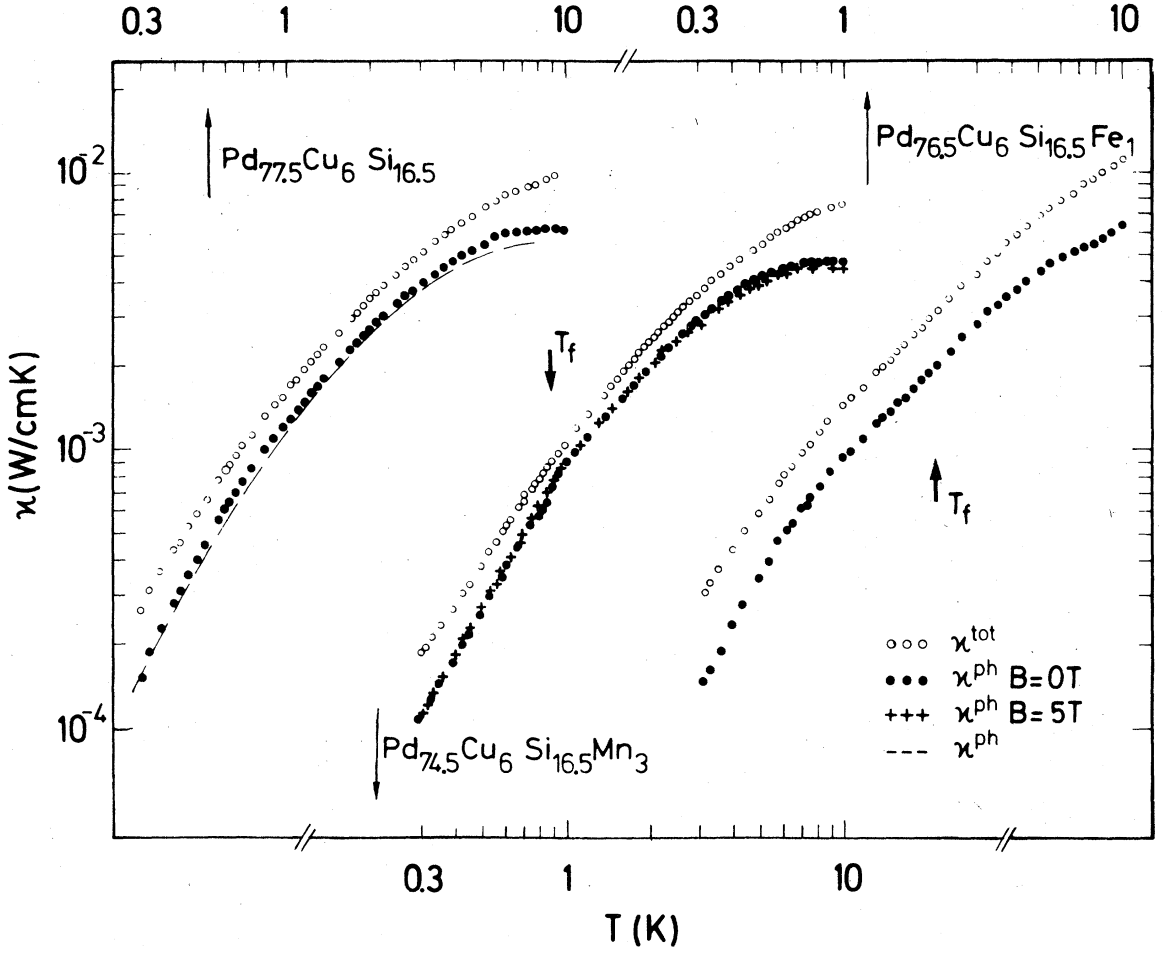

FIG. 1. Thermal conductivity κ of amorphous $Pd_{77.5}Cu_6Si_{16.5}$, $Pd_{74.5}Cu_6Si_{16.5}Mn_3$, and $Pd_{76.5}Cu_6Si_{16.5}Fe_1$ versus the logarithm of the temperature T. Open circles: total conductivity $\kappa^{tot}(T)$; solid circles: phonon thermal conductivity in zero magnetic field $\kappa^{ph}(T)$. Crosses: $\kappa^{ph}(T)$ in a field of 5 T; dashed line: $\kappa^{ph}(T)$ for PdCuSi taken from Ref. 5. The arrows mark the spin-glass freezing temperature of the magnetic alloys.

TABLE I. *(Continued).*

Phonon thermal conductivity ($T \leq 1$ K) $\kappa^{ph} = \alpha T^{\beta}$		Total thermal conductivity at 1 K κ^{tot}(W/cm K)	$\frac{\kappa^{ph}(1\text{ K})}{\kappa^{tot}(1\text{ K})}$ (%)
α (W/cm $K^{1+\beta}$)	β		
1.44×10^{-3}	1.75	1.78×10^{-3}	80.9
1.16×10^{-3}	1.75	1.47×10^{-3}	78.9
8.92×10^{-4}	1.75	1.13×10^{-3}	78.9

scale for the PdCuSi matrix, the PdCuSi + 3 at. % Mn alloy, and the PdCuSi + 1 at. % Fe alloy. Since the samples exhibit a high residual resistivity ρ_0 (see Table I) we assume the validity of the Wiedemann-Franz law (WFL) throughout the investigated temperature range and calculate the phonon contribution κ^{ph} to the thermal conductivity via

$$\kappa^{ph} = \kappa^{tot} - \frac{L_0 T}{\rho_0} \tag{2}$$

with $L_0 = 2.45 \times 10^{-8}$ W Ω K^{-2}, the Lorenz number for free electrons, in agreement with the experimentally determined value $L = 2.4 \times 10^{-8}$ W Ω K^{-2} for PdCuSi at room temperature.[16] The resulting phonon conductivities $\kappa^{ph}(T)$ are also shown in Fig. 1 (solid circles). In addition, the result of Matey and Anderson[5] for a PdCuSi rod of the same composition is given for comparison (dashed line). Taking into account the errors due to the uncertainties in the geometry factor, the results obtained for PdCuSi in different laboratories are quantitatively in good agreement.

Figure 1 clearly reveals that the addition of 3 at.% Mn to PdCuSi qualitatively does not alter the temperature dependence of the phonon thermal conductivity. In both alloys we observe the same features for $\kappa^{ph}(T)$, typical for many insulating and metallic, nonmagnetic and magnetic amorphous materials: a plateau above ~6 K, and a well-known $\kappa^{ph} \propto T^{\beta}$ law for $T \leq 1$ K, with β values of ~1.75, and κ^{ph}(1 K) around 1×10^{-3} W /cm K. Detailed values are given in Table I.

For the 1 at. % Fe sample the same is true with the restriction that for $T \gtrsim 8$ K apparently no plateau occurs. This could be due to enhanced phonon-electron scattering in this sample. That phonon-electron scattering can lead to a reduction or even elimination of the plateau has recently been shown for the amorphous superconductor Zr_7Cu_3.[17] Another possibility is that the Fe sample contains very small fractions which are crystalline, probably indicated by the RRR lying above unity (see Table I). Therefore, contributions from Debye-type phonons in the crystalline regions shunt $\kappa^{ph}(T)$ in the temperature range where normally the plateau occurs.

In Fig. 1 the spin-glass freezing temperatures T_f, as determined from low-field susceptibility measurements, are marked with arrows. One can see that there is no influence of spin freezing and/or cluster blocking in the thermal conductivity around T_f in zero magnetic field.

A further important result of the present work is the absence of any field dependence—at least within the scatter of the data—even in fields as high as 5 T in the heat conductivity of the magnetic samples. This can be seen in Fig. 1 for $\kappa^{ph}(T)$ for the 3 at. % Mn alloy (crosses). The same result is found for a sample containing 5 at. % Fe. A similar observation has been reported earlier for an Fe-Ni sample in low magnetic fields[5] ($B \approx 10$ mT), as well as in a nonmetallic soda-lime glass containing 0.5 wt. % Fe_2O_3 in fields up to 5 T.[12] This means that at least within the accuracy limit in our spin-glass samples the phonons do not scatter from diffuse spin waves that probably exist.

Although there is obviously no additional phonon scattering due to magnetic excitations in PdCuSi, we observe—when comparing absolute values of the conductivities—that on the average κ^{ph} is roughly 30% lower in the alloy containing 3 at. % Mn and about 20% lower in the 1 at. % Fe sample than in the PdCuSi matrix.

It should be stressed that in this investigation one can compare absolute values of the phonon thermal conductivities [and not just superimpose the $\kappa^{ph}(T)$ curves of different samples] because (i) the error due to the geometry factor is small, below ±7%, and (ii) the electronic contribution κ^{el} does not exceed the phonon conductivity κ^{ph} in the entire temperature

range investigated.

In a search for the origin of these reduced conductivities in the magnetic samples we have plotted in Fig. 2 κ^{ph} of the matrix and the Mn alloy (solid symbols), the electronic parts κ_{el} determined from the WFL (dashed lines), and the difference (triangles):

$$\Delta\kappa^{ph} = \kappa^{ph}(\text{matrix}) - \kappa^{ph}(\text{Mn3}) \quad . \tag{3}$$

Figure 3 shows the equivalent plots for the 1 at. % Fe sample.

Obviously in both cases the temperature dependences of $\Delta\kappa^{ph}$ closely resemble those of κ^{ph} itself. From this we infer that the phonon mean free path in the Mn and Fe sample is limited by the same type of scattering centers already present in the matrix.

To further analyze the phonon thermal conductivity within the TLS model we can write

$$(\kappa^{ph})^{-1} = W_{el}^{ph} + W_{TLS}^{ph} \quad , \tag{4}$$

where W_{el}^{ph} is the thermal resistivity due to phonon-electron scattering and W_{TLS}^{ph} the thermal resistivity due to scattering of phonons from TLS. The validity of Eq. (4) implies that the phonons are scattered independently from electrons and TLS (Matthiessen's rule).

Since PdCuSi alloys are not superconducting it is difficult to estimate values of W_{el}^{ph} directly. From our recent investigations[17] on the amorphous superconductor Zn_7Cu_3 it is, however, certain that $\kappa_{el}^{ph} = (W_{el}^{ph})^{-1}$ is a linear function of temperature: $\kappa_{el}^{ph} = D^{-1}T$ for $T < 1$ K, with $D = 2.1 \times 10^3$ K^2 cm/W. A linear temperature dependence of $\kappa_{el}^{ph}(T)$ was also found by comparison of $\kappa^{ph}(T)$ of various amorphous systems, including PdCuSi, where electron-phonon scattering is estimated to be small: $D \approx 1.7 \times 10^2$ K^2 cm/W.[18] Taking this value, at 0.3 K one finds $W_{el}^{ph}/(\kappa^{ph})^{-1} \approx 0.1$. Thus it seems unlikely that the difference $\Delta\kappa^{ph}$ observed after alloying the PdCuSi matrix with Mn or Fe is caused by a change of the electron-phonon scattering. We therfore disregard W_{el}^{ph} in the further dis-

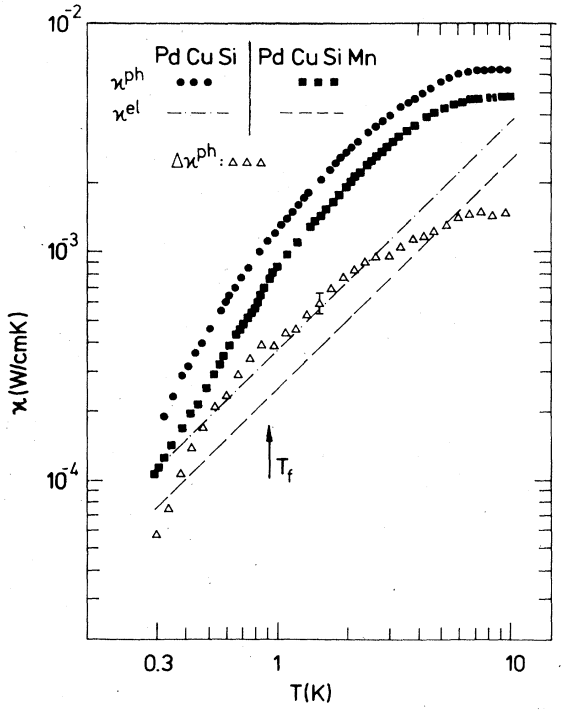

FIG. 2. Phonon thermal conductivity $\kappa^{ph}(T)$ of $Pd_{77.5}Cu_6Si_{16.5}$ (solid circles), $\kappa^{ph}(T)$ of $Pd_{74.5}Cu_6Si_{16.5}Mn_3$ (solid squares), and the difference $\Delta\kappa^{ph} = \kappa^{ph}(\text{PdCuSi}) - \kappa^{ph}(\text{PdCuSiMn})$ (open triangles) versus temperature. The dashed-dotted and the dashed line refer to the electronic parts of the conductivities of κ^{tot} as calculated from the Wiedemann-Franz law of the matrix and the magnetic alloy, respectively.

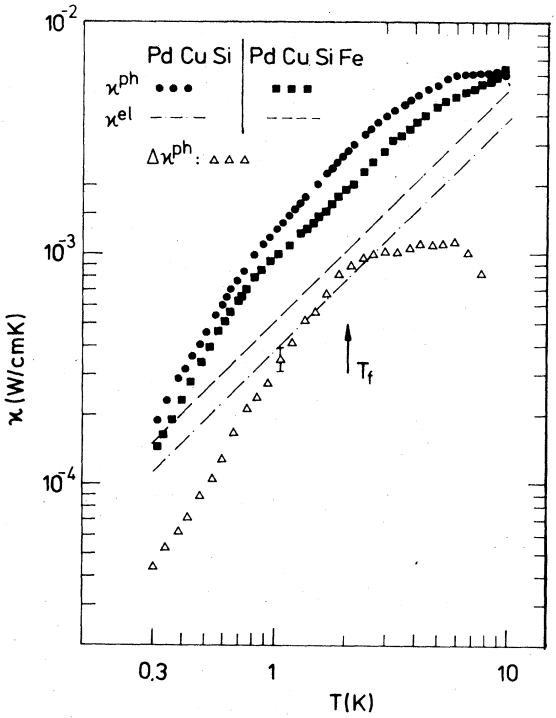

FIG. 3. Same plot as in Fig. 2 for a $Pd_{76.5}Cu_6Si_{16.5}Fe_1$ sample. Solid circles: $\kappa^{ph}(T)$ of the PdCuSi matrix; solid squares: $\kappa^{ph}(T)$ for PdCuSiFe; open triangles: $\Delta\kappa^{ph}(T) = \kappa^{ph}(\text{PdCuSi}) - \kappa^{ph}(\text{PdCuSiFe})$; dashed-dotted and dashed line: electronic contributions to κ^{tot} as calculated from the Wiedemann-Franz law for PdCuSi and PdCuSiFe, respectively.

cussion of $\Delta\kappa^{ph}$ and are left with the term W_{TLS}^{ph} in Eq. (4). Within the TLS model, we can write for the resonant scattering of the phonons from the TLS below ~ 1 K

$$(W_{TLS}^{ph})^{-1} = \kappa_{TLS}^{ph} = 9.87\frac{\rho k_B^3}{6\pi^3\hbar^2}\left[\frac{v_l}{n_0 M_l^2} + \frac{2v_t}{n_0 M_t^2}\right]T^2, \quad (5)$$

where ρ is the density of the material, $v_{l,t}$ the sound velocity in longitudinal and transverse direction, and $n_0 M_{l,t}^2$ the product of the density of the TLS and the respective coupling constants.

If we use ρ as determined experimentally and values for $v_{l,t}$ and $n_0 M_{l,t}^2$ for PdSi as quoted by Belessa,[19] we calculate

$$\kappa_{TLS}^{ph} = 4.4 \times 10^{-3}\ \mathrm{T^2\,W\,cm^{-1}\,K^{-3}}$$

as compared to

$$\kappa_{TLS}^{ph} = 1.4 \times 10^{-3}\ \mathrm{T^2\,W\,cm^{-1}\,K^{-3}}\ ,$$

as determined experimentally for PdCuSi. Similar differences are observed for other amorphous metallic alloys.[5] This shows that the TLS model gives only a qualitative description of the physical processes dominating the low-temperature phonon thermal conductivity.

Nevertheless, an attempt to describe the experimentally observed differences of κ^{ph} between the nonmagnetic matrix and the magnetic alloys leads to the following consequence: Assuming equal coupling constants in Eq. (5) for the matrix and the magnetic alloys, n_0, the density of the TLS, is enhanced after adding Mn and Fe to PdCuSi. Since we do not find any influence of a magnetic field on κ^{ph}, we have no reason to believe that this enhancement is due to the presence of extrinsic magnetic TLS. Ultrasonic attenuation measurements are of no help, since thereby only the product $n_0 M^2$ can be determined, as in the thermal conductivity investigations. The only separate determination of n_0 is possible from measurements of the specific heat. However, in the magnetic samples—as mentioned above—the dominating contributions originate from the internal degrees of freedom of the impurity spins.

IV. SUMMARY AND CONCLUSIONS

We have shown that the addition of a few percent of 3*d* magnetic impurities to amorphous PdCuSi does not alter the temperature dependence of the thermal conductivity at low temperatures. However, we do find an enhanced scattering of phonons in the magnetic alloys as compared to the diamagnetic PdCuSi matrix. From this we infer within the two-level system model an enhanced density of TLS. The absence of any effects in $\kappa^{ph}(T)$ of either spin freezing (or cluster blocking) at the spin-glass freezing temperatures T_f or of external magnetic fields leads to the conclusion that the scattering enhancement is not of magnetic origin. Therefore, the results of these experiments do not give evidence for the existence of extrinsic magnetic TLS in the investigated alloys. It remains an open question why the concentration of TLS in the amorphous alloys is higher after adding 3*d* elements. If the TLS are microscopically identified with voids as, e.g., recently discussed by Banville and Harris[20] one would deduce that the magnetic alloys are less dense than PdCuSi, since the more open a glassy structure, the greater the phonon scattering and thus the greater the density of TLS. We find, however, from our density measurement that ρ of PdCuSi does not differ by more than 1% from ρ of the magnetic samples. This is in accordance with measurements by Chen and Park,[21] who found only little changes ($\sim 2\%$) in ρ when varying the Cu and Si concentration in PdCuSi. This means that from density measurements alone one cannot find a conclusive answer.

One possibility for changing the density is to anneal the samples just below the glass temperature, thereby relaxing the structure, and at the same time reduce the concentration of TLS. We have done this for the PdCuSi and (PdCuSi)Mn samples and find an increase in κ^{ph} between 15% and 30%, similar to the results obtained by Matey and Anderson.[5] The difference in κ^{ph} between the matrix and the magnetic sample is, however, not diminished appreciably. From this we infer that possibly the addition of Mn stabilizes the glassy state of PdCuSi and enhances the glass temperature, similar to the effect of Cu addition to PdSi.[22] Further investigations on a larger variety of samples are under way to lead to a better understanding of these problems.

Note added in proof. From a recent paper by W. Arnold, P. Doussineau, Ch. Frenois, and A. Levelut [J. Phys. (Paris), in press] and a private communication from S. Hunklinger we learned that at least in superconducting Zr_7Pd_3 and Nb_2Zr_8 values for $n_0 \times M_{l,t}^2$ as deduced from ultrasonic attenuation measurements are a factor of 2—4 higher than originally assumed. If the same is true for PdSi and PdCuSi, i.e., that an equally large in-

crease in $n_0 \times M_{l,t}^2$ follows from the ultrasonic attenuation or are metallic glasses, the observed difference in κ_{TLS}^{ph} when calculated from Eq. (5) and the value we found experimentally is completely removed. As a consequence, the TLS model also gives a good *quantitative* description of $\kappa^{ph}(T)$. We thank S. Hunklinger and W. Arnold for bringing this point to our attention.

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