Structural and magnetic ordering effects in $(Tb_{1-x}Y_x)Cu$ systems

LETTER TO THE EDITOR

Structural and magnetic ordering effects in $(Tb_{1-x}Y_x)Cu$ systems

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Abstract. Structural transformation and magnetic ordering effects have been observed at temperatures T_S and T_N, respectively, in polycrystalline as well as in single-crystalline samples of $(Tb_{1-x}Y_x)Cu$. For the TbCu ($x = 0$) system, the values of T_S and T_N are found to be the same for the polycrystalline sample, whereas they differ for the single-crystalline one. Furthermore, for all values of x, the value of T_N is the same for both the above classes of samples. However, the value of T_S for the single-crystalline samples is found to be larger by about 10–20 K compared to that for the polycrystalline ones.

The understanding of both structural and magnetic behaviour has been the subject of interest [1–9] in several RCu (R = Y, La to Lu) compounds. An important question [2] is whether the structural transformation and whether the antiferromagnetic (AF) behaviour arises from a single mechanism or not. For GdCu, the structural transition temperature, T_S, has been found to be different from the AF ordering temperature, T_N, whereas in TbCu, it was found that the above two temperature scales are almost of the same value [1–3]. Furthermore, thermal hysteresis effects around T_S have also been observed [2, 7, 10] in many RCu compounds. For example; in the TbCu compound, a large orthorhombic (FeB type) distortion in its cubic (CsCl) type structure sets in [3] at temperatures below T_S ($\simeq 117$ K).

Two models have been suggested for the lattice transformation effects, at T_S, in the RCu class of compounds. The first one [5] attributes it to Jahn–Teller (J–T) effects. The J–T effects give rise to the splitting of the d band of the R atom. This leads to a large variation in the density of states (DOS) at E_F. The second model suggests [2] that quadrupolar interactions could also be responsible for inducing the structural transition from the cubic to non-cubic phase of the RCu compounds. In the cubic structure there is no electric field gradient (EFG) and hence quadrupole moments of the R atoms would have no interaction [1, 2] with the lattice. Therefore, they would be randomly oriented. However, in the non-cubic (orthorhombic) structure, a finite EFG would align the quadrupole moments. This would cause strains which would lead to a structural transition.

Up to now, there have been very few studies [1, 10] on single-crystalline samples in the above mentioned class of compounds. The study on the YCu and the TbCu class of compounds would enable us to examine the various characteristic temperature scales and the origin of T_S and T_N. In TbCu, a large thermal hysteresis has been observed around $T_S \simeq T_N$. On the other hand, for YCu, the value [3, 4] of $T_S \simeq 140$ K, and no magnetic ordering, have been observed in this system; it behaves like a Pauli paramagnet down to 4.2 K. We have therefore prepared the $(Tb_{1-x}Y_x)Cu$ alloy ($x = 0, 0.1, 0.4, 1$) series, both

in the single-crystalline and in the polycrystalline phase so as to investigate the behaviour of both T_S and T_N with variation of x.

The polycrystalline samples were prepared in an arc furnace under flowing argon conditions. The alloys were repeatedly melted to ensure homogeneity. Rods of 2 mm diameter and 3 cm length were drawn using the cold suction method. These samples were then annealed at 700 °C for one week. All the alloys were found to crystallize in the CsCl type of cubic structure. The lattice spacings (table 1) of all the polycrystalline alloys prepared by us are found to match with the earlier reported values [3, 4].

Table 1. Lattice parameter, a, for the polycrystalline and the single-crystalline samples of the $(Tb_{1-x}Y_x)Cu$ system.
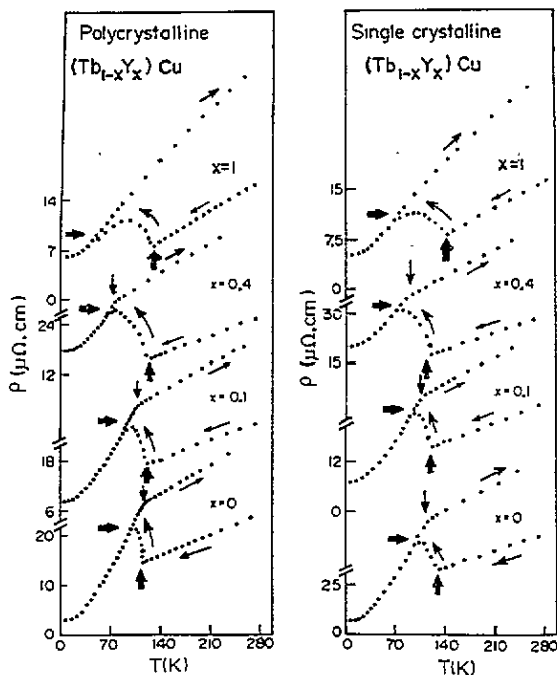
System $(Tb_{1-x}Y_x)Cu$	Polycrystalline a (Å)	Single-crystalline a (Å)
$x = 0$	3.484 ± 0.015	3.486 ± 0.010
0.1	3.486 ± 0.020	3.490 ± 0.009
0.4	3.487 ± 0.016	3.494 ± 0.012
1.0	3.489 ± 0.010	3.497 ± 0.008

A triarc single-crystal puller (Centorr Furnaces, USA) was employed to prepare single-crystalline samples using the Czochralski technique. The single crystals were grown on a cold tungsten tip, connected to a heat pipe. The pulling and the rotational speeds of the tungsten tip were 10 mm h^{-1} and 10 revolutions per minute (RPM), respectively. The melt was rotated in a graphite crucible with a speed of 10 RPM, in a direction opposite to that of the tungsten tip. The values (table 1) of the lattice spacings of the single-crystalline samples are close to those for the polycrystalline ones.

The resistivity, ρ, data reveal (figure 1) complex behaviour at T_S and T_N. As the samples are cooled below the transition temperature, T_S (thick vertical arrows pointing upwards (figure 1)), a sudden jump is observed in the ρ curves similar to the earlier reported study of the YCu ($x = 1$) compound. Below T_S, the ρ curves rise rapidly up to a temperature T_H (thick horizontal arrow) whose value (table 2) is about 20–80 K smaller than that for T_S. T_H indicates the temperature where the structural transformation process is completed. For $T_H \leqslant T \leqslant T_S$, the cubic phase gradually transforms [3] to the low-temperature orthorhombic phase, as the temperature is lowered. In the fully transformed orthorhombic phase ($T < T_H$), the heating and the cooling curves are identical (figure 1). For $T > T_H$, a large thermal hysteresis is observed (figure 1) for all the alloys. At the AF transition temperature, T_N (thin vertical arrows pointing downwards (figure 1)), the ρ curves (figure 1) show a clear change in slope. The fine arrows (figure 1) indicate the ρ curves as exhibited during the cooling and heating cycles. For the polycrystalline TbCu ($x = 0$) system, we measured $T_N = T_S = 118$ K, whereas for the single-crystalline system, although the value of T_N is the same as the polycrystalline sample, the value of T_S (table 2) increases to 130 K. With increase in x, the T_N value (table 2) drops. The most important observation is that for all values of x, the value of T_S for the single-crystalline samples is greater (table 2) than that for the corresponding polycrystalline samples. This could be due to the fact that for the single-crystalline samples, the lattice imperfections are fewer in number compared to the polycrystalline samples. The onset of the cubic–orthorhombic structural transformation at T_S could thus become energetically favourable at relatively higher temperatures.

Table 2. Characteristic temperature scales, as obtained from our ρ studies, for the polycrystalline and the single-crystalline samples of the $(Tb_{1-x}Y_x)Cu$ system.

System	Polycrystalline			Single-crystalline		
$(Tb_{1-x}Y_x)Cu$	T_S (K)	T_N (K)	T_H (K)	T_S (K)	T_N (K)	T_H (K)
$x = 0$	118	118	105	130	118	103
0.1	121	108	85	125	108	103
0.4	120	78	60	130	78	70
1.0	126	—	40	145	—	60

Figure 1. ρ versus T curves, for the polycrystalline and single-crystalline samples of the $(Tb_{1-x}Y_x)Cu$ system. The thick horizontal arrow denotes T_H. The thick vertical arrow (pointed upwards) indicates T_S. The thin vertical arrow (pointed downwards) indicates T_N. All other arrows indicate different temperature cycles (heating and cooling) employed.

The thermopower, S, (figure 2) and the thermal conductivity, λ, (figure 3) also show an abrupt jump at T_S and large thermal hysteresis effects for $T > T_S$. The values of T_S and T_H from these measurements also agree with those observed in the ρ measurements. In all the S curves, for both the polycrystalline and the single-crystalline samples, the high-temperature $(T > T_S)$ cubic phase has a value of $S \sim 10~\mu V~K^{-1}$ at 300 K, whereas the orthorhombic phase has a smaller value of $S \sim 4$–5 $\mu V~K^{-1}$. The λ curves also exhibit a higher value in the cubic phase than that for the orthorhombic phase. For $T < T_S$, the S curve (figure 2) exhibits a prominent maximum (for $x = 0$) at $T_{max}^S = 30$ K. This maximum gradually broadens out, with an increase in x, and finally is obliterated (figure 2) around $x \sim 0.4$. The λ curve (figure 3) also exhibits a clear maximum (for $x = 0$) at $T_{max}^\lambda = 35$ K. This maximum is also gradually obliterated, with increasing x, and instead exhibits (figure 3) a

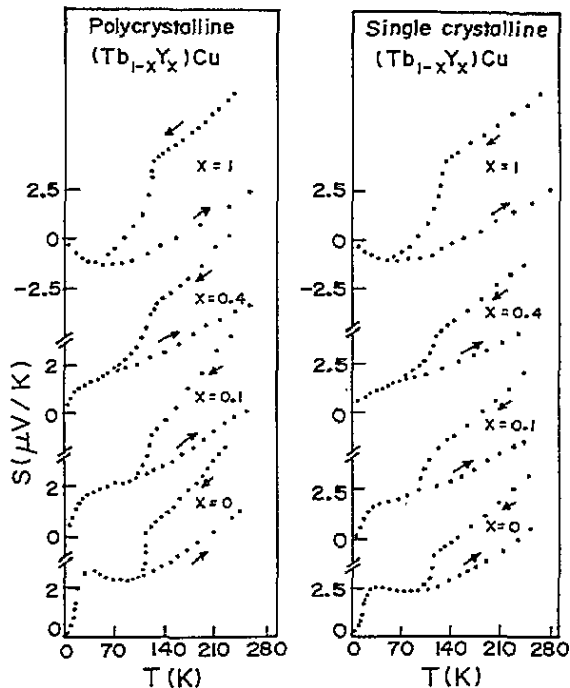**Figure 2.** S versus T curves for the polycrystalline and single-crystalline samples of the $(Tb_{1-x}Y_x)Cu$ system. The arrows indicate different temperature cycles (heating and cooling) employed.

change in slope at $T < 30$ K. Such extremum features in the S and λ behaviours have been observed [11–13] earlier in AF compounds like RCu_2 (R = Tb, Sm) and in ferromagnetic compounds such as RAl_2 (R = Sm, Tb, Gd). The above features in our systems could thus arise from spin waves below T_N. They act as an additional channel for heat conduction and electron scattering and give rise [11–13] to an extra contribution to λ and S. This could account for the observed maximum features (figures 2 and 3) below T_N. With increasing x, as $T_N \rightarrow 0$, these extremum features were found (figures 2 and 3) to be suppressed.

We now make a few comments on the physical origin for the occurrence of the temperature scales T_S and T_N. In YCu ($x = 1$), the sudden increase in the ρ curve (figure 1) for $T < T_S$ has been attributed [5,7] to the large variation in the DOS due to J–T effects. Similar ρ behaviour has been observed for all our $(Tb_{1-x}Y_x)Cu$ systems also. The observation of a sudden drop in the S and the λ curves (figures 2 and 3), for $T < T_S$, could also thus indicate a sharp change in the DOS. This seems to indicate that the structural transformation effect in our systems, over the entire alloy series, could arise due to the J–T type of distortion.

We now discuss the suggestion [2] that the quadrupole interactions could also be the driving force for the AF as well as for the structural transition. We note that quadrupole moments, in the presence of CF effects, could give rise [14] to an S-shaped behaviour in the ρ curve, at $T \sim 100$ K, with a tendency to saturate at high temperatures ($T \sim 300$ K). Such behaviour has been observed [14] earlier in compounds such as $PrPb_3$, because of which the structural transformation effects have been attributed to quadrupolar effects. However, in all our systems for $T > T_S$, the ρ curves (figure 1) exhibit an approximately linear
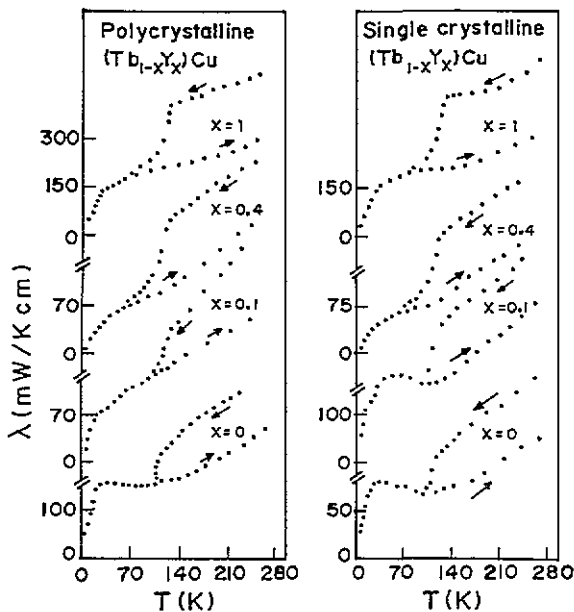
Figure 3. λ versus T curves for the polycrystalline and single-crystalline samples of the $(Tb_{1-x}Y_x)Cu$ system. The arrows indicate different temperature cycles (heating and cooling) employed.

behaviour for both the cubic and the orthorhombic phases. Such linear behaviour could arise by simply considering the usual electron–phonon interaction. Our ρ results, therefore, seem to indicate the absence of quadrupole induced lattice transformation effects in the $(Tb_{1-x}Y_x)Cu$ systems.

The most important result from our studies is the observation that, for all values of x, T_S for the single-crystalline sample is greater, by about 10–20 K, than that measured for the polycrystalline samples. The value of T_S is thus found to depend on the nature of sample preparation, whereas T_N does not. Further, in the $(Tb_{1-x}Y_x)Cu$ system, the structural transition effect could arise from the Jahn–Teller type of strain induced in the lattice.

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