Evolution of a metastable phase with a magnetic phase coexistence phenomenon and its unusual sensitivity to magnetic field cycling in the alloys $\text{Tb}_{5-x}\text{Lu}_x\text{Si}_3$ ($x \leq 0.7$)

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Abstract

Recently, we reported an anomalous enhancement of the positive magnetoresistance beyond a critical magnetic field in $\text{Tb}_5\text{Si}_3$ in the magnetically ordered state, attributable to ‘inverse metamagnetism’. This results in unusual magnetic hysteresis loops for the pressurized specimens, which are relevant to the topic of ‘electronic phase separation’. In this paper, we report the influence of small substitutions of Lu for Tb, to show the evolution of these magnetic anomalies. We find that, at low temperatures, the high-field high-resistivity phase could be partially stabilized on returning the magnetic field to zero in many of these Lu substituted alloys, as measured through the electrical resistivity ($\rho$). Also, the relative fractions of this phase and the virgin phase appear to be controlled by a small tuning of the composition and temperature. Interestingly, at 1.8 K a sudden ‘switch-over’ of the value of $\rho$ for this mixed phase to that for the virgin phase for some compositions is observed at low fields after a few field cycles, indicating metastability of this mixed phase.

1. Introduction

The compounds exhibiting metamagnetic transitions and ‘electronic phase separation’ (EPS) have been attracting a lot of attention in condensed matter physics in recent years [1]. An intermetallic compound, $\text{Tb}_5\text{Si}_3$ (with a Neél temperature of $T_N \sim 70$ K), and its derivatives have been recently identified by us as behaving exceptionally in these respects. A strong enhancement of the positive magnetoresistance (MR, defined as $(\rho(H) - \rho(0))/\rho(H)$) at a critical magnetic field ($H_c$), instead of a decrease of the MR (characterizing ‘metamagnetism’), is observed in these alloys [2]. This anomaly was attributed to ‘inverse metamagnetism’—a concept not so commonly known among metallic magnets—in which ‘magnetic fluctuations’ instead of ‘ferromagnetic alignments’ are favored [1, 3] beyond $H_c$. During the course of the investigation of $\text{Tb}_5\text{Si}_3$ under external and chemical pressure [4–6], we made many important findings bearing relevance to the concept of EPS. For the parent compound, under ambient pressure conditions, though a strong irreversibility is observed in the plots of the isothermal magnetization ($M$) and MR versus $H$, the curve obtained while reversing the field merges with the virgin curve before attaining zero field. However, under high pressure (e.g., 10 kbar) and for $\text{Tb}_5\text{Lu}_3\text{Si}_3$, the high-field phase is at least partially retained after returning the field to zero, giving rise to an unusual EPS comprising a high-field high-resistivity phase and a low-field low-resistivity phase [5, 6]. Thus, this alloy is characterized by an MR($H$) hysteresis loop that is uncommon in magnetism, as the virgin curve lies below the envelope loop in the plot of MR versus $H$. This unique feature provided us with the basis for the present investigation of the alloy series $\text{Tb}_{5-x}\text{Lu}_x\text{Si}_3$, for small doping of Lu ($x < 1$), allowing us to explore more carefully how such unusual MR loops evolve.

2. Experimental details

Polycrystalline samples, $\text{Tb}_{5-x}\text{Lu}_x\text{Si}_3$ ($x = 0.1, 0.3, 0.5$ and 0.7), were prepared by arc melting stoichiometric amounts.
implies that the high-field phase is partially stabilized [7–9] as compared to cases for previous concentrations. Also small drops/jumps are observed in cycles 2, 4 and 6 near 7, 8 kOe, implying that the virgin state can be attained after repeated field cycling, indicating metastability of the phase before cycle 5 begins. Such a drastic drop in \( \rho \) has been obtained in the parent compound Tb\( _5 \)Si\( _3 \) by applying external pressure—however, also by a small application of magnetic field for the sample series Tb\( _{1−x} \)Lu\(^{x} \)Si\( _3 \) with \( x = 0.1, 0.3, 0.5 \) and 0.7 at 1.8 K. ((e), (f)) Isothermal magnetization for the same sample series at 1.8 K. Arrows and numerics are given as a guide to the eyes. The inset shows electrical resistivity as a function of temperature for all the alloys to highlight the \( x \)-dependence of the magnetic transition temperature.

This figure is in colour only in the electronic version.

3. Results and discussion

Figures 1(a)–(d) show the magnetic field response of MR (left panel) and \( M \) (right panel) at 1.8 K for all the compositions for six cycles (cycle 1: 0–120 kOe; cycle 2: 120–0 kOe; cycle 3: 0–120 kOe; cycle 4: −120–0 kOe; cycle 5: 0–120 kOe; cycle 6: 120–0 kOe). We have also performed measurements of \( \rho \) as a function of temperature for all the compositions to demonstrate the gradual reduction in the onset of the magnetic ordering temperature (from \( \sim 70 \) K for \( x = 0 \) to \( \sim 55 \) K for \( x = 0.7 \)) as indicated by the temperature where \( \rho \) falls (see figure 1, inset). We have not shown the data for the parent compound here, as these were discussed in our earlier publications [2, 4] (and are also similar in features to those for \( x = 0.1 \), presented below). From figure 1(a), it is obvious that, for the \( x = 0.1 \) composition, the field where the steep rise in MR is observed (near 55 kOe) coincides with the field where a jump is observed in the \( M(H) \) curve in cycle 1. Beyond this critical field, MR falls gradually with \( H \) and rises with decreasing field in cycle 2. There is a drastic fall in \( \rho \) (\( \sim 29 \) kOe) and \( M \) (\( \sim 19 \) kOe) in cycle 2 implying that the reverse transition is complete within the first quadrant itself. (The critical fields inferred through these two techniques marginally differ, which we attribute to different responses to local inhomogeneities due to the Lu substitution.) The features observed in cycles 3 and 4 are essentially mirror images of those for the first quadrant. A rise as in cycle 1 and a fall similar to that of cycle 2 are also observed in cycles 5 and 6 respectively at almost the same fields. These features are similar to those observed for the parent compound [2, 4]. For \( x = 0.3 \), as seen in figure 1(f), the shape of the \( M(H) \) curve is similar to that for \( x = 0.1 \), showing jumps/drops at the respective cycles with a marginal decrease in the critical field values. However, in MR(\( H \)) (figure 1(b)), even though an enhancement in resistivity is observed in the first cycle (following the \( M(H) \) behavior), no drastic fall is observed in cycle 2 while reducing the field in the first quadrant. This implies that the high-field phase is partially stabilized [7–9] at \( H = 0 \). As argued earlier [4–6], in the high-field phase, magnetic fluctuations are introduced and the magnitude of MR for this phase smoothly reduces (cycle 2) with \( H \) (essentially quadratically, which favors persistence of these fluctuations, as discussed in [6]) as \( H \) is reversed towards zero. The scattering influence of this high-field phase persists in cycle 3 as well (figure 1(b)), but, in cycle 4, a drastic drop in MR is observed near \( −8 \) kOe, implying that the virgin state can be attained after repeated field cycling, indicating metastability of the phase before cycle 5 begins. Such a drastic drop in \( \rho \) has been obtained in the parent compound Tb\( _5 \)Si\( _3 \) by applying external pressure—however, also by a small application of magnetic field in the negative direction [6]. The transformation to the virgin phase is substantiated from the observation that the curves in cycles 5 and 6 are of the same type as those in cycles 1 and 2.

Focusing on the magnetic behavior (figure 1(c)) of the composition \( x = 0.5 \), there is a subtle difference with respect to that for \( x = 0.3 \). In \( M(H) \), the transition in cycle 1 is somewhat more broadened with a jump of a lower magnitude, as compared to cases for previous concentrations. Also small drops/jumps are observed in cycles 2, 4 and 6 near 7, \( −6.2 \) and 5.8 kOe respectively. As regards the MR(\( H \)) curve of this compound, the features are similar to those for \( x = 0.3 \) in

of high-purity (>99.9 wt%) constituent elements in an atmosphere of high-purity argon. The single-phase nature of the specimens was ascertained by means of x-ray diffraction (Cu K\( \alpha \)). With the increase in composition of Lu, the volume of the compound is, as expected, found to decrease (for \( x = 0.1 \); \( a = 8.437 \) Å, \( c = 6.341 \) Å, \( V = 390.95 \) Å\(^3 \) and for \( x = 0.7 \); \( a = 8.427 \) Å, \( c = 6.335 \) Å, \( V = 389.60 \) Å\(^3 \), with a typical error of \( ±0.004 \) Å in \( a \) and \( c \), establishing an increase of positive chemical pressure. The measurements of \( \rho \) as a function of the magnetic field at 1.8 and 5 K (<120 kOe) were performed using a commercial physical property measurements system (Quantum Design) and a conducting silver paint was used for making electrical contacts of the leads with the samples. In addition, dc \( M \) measurements at 1.8 and 5 K (<120 kOe) were performed with the help of a commercial vibrating sample magnetometer (Oxford Instruments), for a comparative investigation.

Figure 1. ((a)–(d)) MR as a function of externally applied magnetic field for the sample series Tb\( _{1−x} \)Lu\(^{x} \)Si\( _3 \) with \( x = 0.1, 0.3, 0.5 \) and 0.7 at 1.8 K. ((e), (f)) Isothermal magnetization for the same sample series at 1.8 K. Arrows and numerics are given as a guide to the eyes. The inset shows electrical resistivity as a function of temperature for all the alloys to highlight the \( x \)-dependence of the magnetic transition temperature.
cycles 1–3. However, in cycle 4, while, for the composition $x = 0.3$, the high-field phase becomes unstable as one approaches zero field, for $x = 0.5$, this high-field phase persists until $H = 0$ in this cycle. The nature of cycle 5 is also different from that for the other compositions ($x < 0.5$), in the sense that a gradual downturn is observed with increasing $H$ (for $x = 0.5$), merging with the virgin curve beyond $H_c$ only. Interestingly, in the sixth cycle, a drastic drop in $\rho$ is observed near 2 kOe, returning the $\rho$ value to that of the virgin state, implying that the ‘switching’ behavior is shifted to the near end of cycle 6. If one looks at the $MR(H)$ behavior of the composition $x = 0.7$, no drop is observed in any cycle (other than that known for the virgin curve), and the high-field phase persists in the vicinity of $H = 0$ (with a monotonic decrease of the magnitude of $MR$ with increasing $H$) after traveling through the magnetic transition in cycle 1. The envelope curve in the $MR$ plot lies above the virgin curve like that for $x = 1.0$ [5]. Below 70 kOe, $M$ decreases linearly with $H$ in the reverse cycle in the first quadrant, similar to that for a paramagnet, without any evidence for a first-order transition, which supports the idea of increasing dominance of ‘fluctuations’ in the high-field phase in zero field after cycle 1. The $M(H)$ curve however reveals a weak drop as soon as the field direction is reversed, as though there is a fraction transforming to the virgin phase. At this point we would like to mention that the field at which the drop is observed in $M$ in cycle 2 decreases from 13 to 6 kOe from $x = 0.1$ to 0.5, and this is consistent with the absence of a drop in cycle 2 for $x = 0.7$.

From the discussions above, it can be concluded that the high-field phase that is stabilized in zero field coexists with the virgin phase after cycle 2 for $x > 0.1$ and, qualitatively speaking, viewing this together with the $M(H)$ data, we can say that the fraction of this high-field phase tends to dominate the electrical conductivity with increasing $x$. To give more evidence for the idea of phase coexistence (or electronic phase separation), we performed measurements of $MR(H)$ at a higher temperature, 5 K. Figures 2(a)–(d) show the $MR(H)$ of the four compositions. It is seen that the signature of EPS is absent at $H = 0$ (after cycling across $H_c$) for the composition $x = 0.1$, as the curves fall back to the virgin curves within the first quadrant in cycles 2 and 4. As we move to the concentration $x = 0.3$, it appears as though only a very small fraction of the high-field phase dominates the conductivity at $H = 0$ for this temperature in cycles 2 and 4, as indicated by the $MR$ value which is closer to that of the virgin phase. For $x = 0.5$, a significant fraction of the high-field phase coexisting with the virgin phase dominates the conductivity, as is evident from the (intermediate) value of $MR$ after the second and fourth cycles. Interestingly, for this composition, a small jump is visible close to zero field after cycles 2 and 4, as though there is a tendency towards ‘switching behavior’ even at this temperature. The shape of the entire curve is butterfly-like with the virgin curve lying below the envelope curve. For the highest concentration $x = 0.7$ in the present study, the domination of the high-field phase for the conductivity relative to that of the virgin phase further increases at this temperature. These trends are rather consistent with those observed at 1.8 K. Hence this figure yields clear evidence for the tuning of the ‘mixed phase’ and ‘the dominance of electrical conductivity by a desired phase’, by varying the chemical pressure.

4. Summary

This alloy series provides an opportunity to probe the evolution of an unusual electronic phase separation involving high-field high-resistivity and low-field low-resistivity phases. We have shown that, under ambient pressure conditions, the electronically phase-separated magnetic phase can be tuned to change its electrical resistivity at low fields when subjected to magnetic field cycling. There are jumps in the magnetoresistance in the vicinity of zero field after a field cycling for certain alloys, the origin of which is puzzling. This work thus emphasizes the need to obtain a theoretical understanding of various manifestations of metastable magnetic phases.

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