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Magnetic and resistive properties of $TbFe_2Si_2$

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Abstract. Detailed studies of the magnetic properties and resistivity of the ternary intermetallic compound $TbFe_2Si_2$ are presented. Besides the low-temperature antiferromagnetic transition at $T_N = 6.5$ K, a possible second anomaly in the magnetic and transport properties has been found at about $T_{SC} = 180$ K. Below T_N, a metamagnetic behaviour was observed. In the region $T_N < T < T_{SC}$, the magnetic susceptibility and magnetization data show that an extra spin magnetic moment appears, in addition to the Curie–Weiss and Pauli paramagnetism. It is suggested that the observed anomalies in the temperature dependencies of the resistivity and magnetic susceptibility are related to the effect of spin correlations. Several mechanisms of the spin fluctuations are discussed.

1. Introduction

The rare-earth ternary intermetallic compound $TbFe_2Si_2$ has the $ThCr_2Si_2$-type crystal structure with space group $I4/mmm$ [1]. The dimension of the tetragonal unit cell is approximately $4 \times 4 \times 10$ Å^3. The neutron diffraction study [1] revealed that at low temperatures $TbFe_2Si_2$ transforms from a paramagnetic state into an antiferromagnetic (AF) state, and the Néel temperature T_N is 10.5 K. Only Tb atoms possess a magnetic moment and the magnetic structure is incommensurate with the crystallographic unit cell. The ^{57}Fe Mössbauer spectroscopy measurements by Noakes *et al* [2] have proved that Fe atoms in $TbFe_2Si_2$ possess no magnetic moments, and the Fe atoms occupy only one type of local site, i.e., there is no site exchange of the Fe and Si atoms. The magnetic transition temperature measured by ac susceptibility is about 5.8 K [2]. The values of the Néel temperature determined by two research groups are not consistent, and there are no data on the electric properties of $TbFe_2Si_2$. In this paper we present detailed measurements of the resistivity and magnetic properties of the polycrystalline $TbFe_2Si_2$ compound. Besides the antiferromagnetic transition at 6.5 K, we have found an additional magnetic phase transition at high temperature.

2. Experimental details

Polycrystalline $TbFe_2Si_2$ samples were prepared by the melting of stoichiometric amounts of the constituent materials in an argon arc furnace. The purities of the materials were Tb:3N5, Fe:6N and Si:6N. To improve the homogeneity, the resulting metallic buttons were

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remelted several times after turning them over. During melting the overall weight loss was less than 1%. Then the samples were sealed in a quartz tube and annealed at 800 °C for seven days.

The x-ray measurements were performed with a MAC MXP3 x-ray diffractometer. The x-ray diffraction spectrum of $TbFe_2Si_2$ can be fitted well to a body-centred-tetragonal crystal structure of the $ThCr_2Si_2$ type with space group $I4/mmm$. The unit-cell parameters a and c are 3.925 Å and 10.088 Å, respectively, and they are consistent with the data of reference [1].

The magnetic susceptibility and magnetization measurements were performed using the Physical Property Measurement System (PPMS) (Quantum Design Company), over the temperature range 2–300 K, and in an applied magnetic field H of up to 30 kOe. In all of the measurements, the sample was first cooled down to a specified temperature in zero field, then an external magnetic field was applied, and all of the data were taken at each step of the temperature and field variations. The resistivity measurements were also performed using the PPMS with a standard four-probe technique.

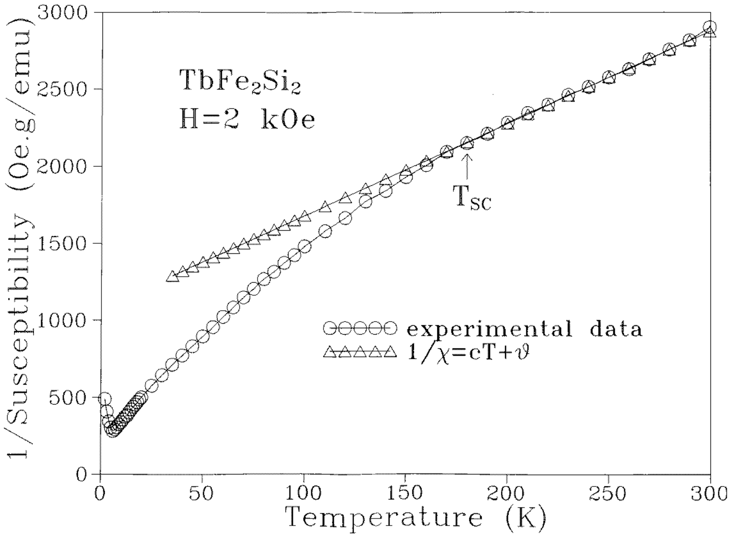

Figure 1. The temperature dependence of the inverse magnetic susceptibility in $TbFe_2Si_2$ measured in an applied field of 2 kOe.

3. Results and analysis

Figure 1 shows the temperature dependence of the inverse susceptibility of $TbFe_2Si_2$ in the field $H = 2$ kOe. Two anomalies are observed in the $1/\chi = f(T)$ dependence. The low-temperature anomaly at 6.5 K is typical of the magnetic phase transition from a paramagnetic into an antiferromagnetic state, and our value of $T_N = 6.5$ K is close to the result of Noakes *et al* [2]. The second anomaly appears at around $T_{SC} = 180$ K as the $1/\chi(T)$ curve deviates from linear behaviour (figure 1). It may be an indication of the second phase transition which has not been found in previous studies.

From the slope of the linear part in the $1/\chi(T)$ curve (between 180 and 290 K) we calculated the value of the magnetic moment $\mu_{eff} = 21.03\ \mu_B$. Since Fe atoms possess no magnetic moment in TbFe$_2$Si$_2$, we have to allow that all of the moment arises from Tb atoms. However, the moment of a free Tb atom is 9.72 μ_B, and it is much lower than the calculated value. In the neutron studies [1], the value of μ_{eff} was not found for TbFe$_2$Si$_2$, but for the similar compounds with the 3d elements Co, Ni and Cu it is in the range 8.6–9.2 μ_B [1].

We found that in the high-temperature range, where the contributions from Tb crystal-field splitting of the ground state may be neglected, the susceptibility can be fitted to the law

$$\chi = \chi_0 + C/(T - \theta)$$

were χ_0 is the temperature-independent contribution due mainly to the itinerant-electron Pauli paramagnetism. The best fit gives the Pauli term $\chi_0 = (2.1\text{–}2.4) \times 10^{-4}$ emu g^{-1} Oe^{-1}, and then the Tb magnetic moment reduces to the reasonable value $\mu_{eff} = 9.23\text{–}7.68\ \mu_B$.

It is seen in figure 1 that at $T < T_{SC}$ the inverse susceptibility deviates downwards from the linear dependence. This implies that below 180 K, an extra spin magnetic moment appears, in addition to the Curie–Weiss and Pauli paramagnetism.

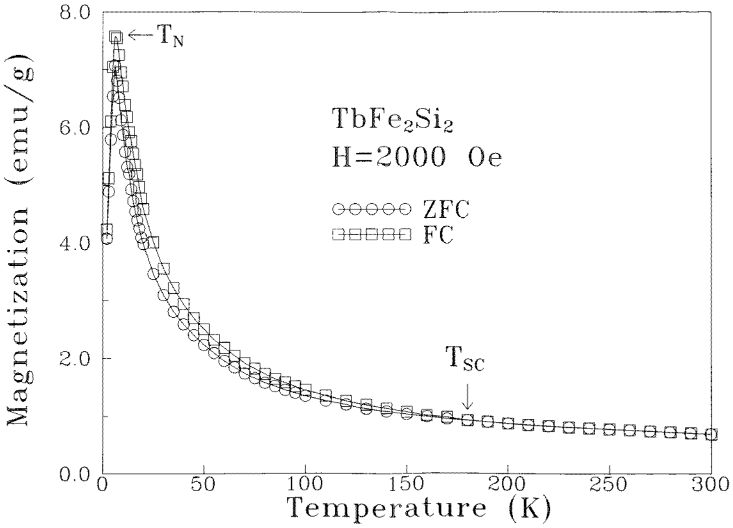

Figure 2. The temperature dependence of the magnetization in TbFe$_2$Si$_2$ for ZFC and FC regimes in the applied field of $H = 2$ kOe.

In order to identify the nature of the extra magnetic moment, the temperature dependence of the magnetization $M(T)$ of TbFe$_2$Si$_2$ was measured over the temperature region 2–300 K in both zero-field-cooling (ZFC) and field-cooling (FC) regimes, and the results are shown in figure 2. We observed that at T_N, the peak positions of the magnetization in the two curves are the same. However, below 180 K, the FC and ZFC curves are split and the FC magnetization is higher than the ZFC one. This is an additional indication of spin correlations at $T < 180$ K. The temperature 180 K is just the same as the T_{SC}-value in the $1/\chi(T)$ curve. The field dependencies of the magnetization $M(H)$ of TbFe$_2$Si$_2$ at the temperatures below and above T_N are shown in figures 3(a) and 3(b), respectively. At no temperature is hysteresis observed. At $T = 2$ K, the magnetization curve is rather unusual (figure 3(a)). With the field increasing, the magnetization increases linearly at low field, then it changes slope several times at about $H = 7$ and 12 kOe, and then tends to saturate

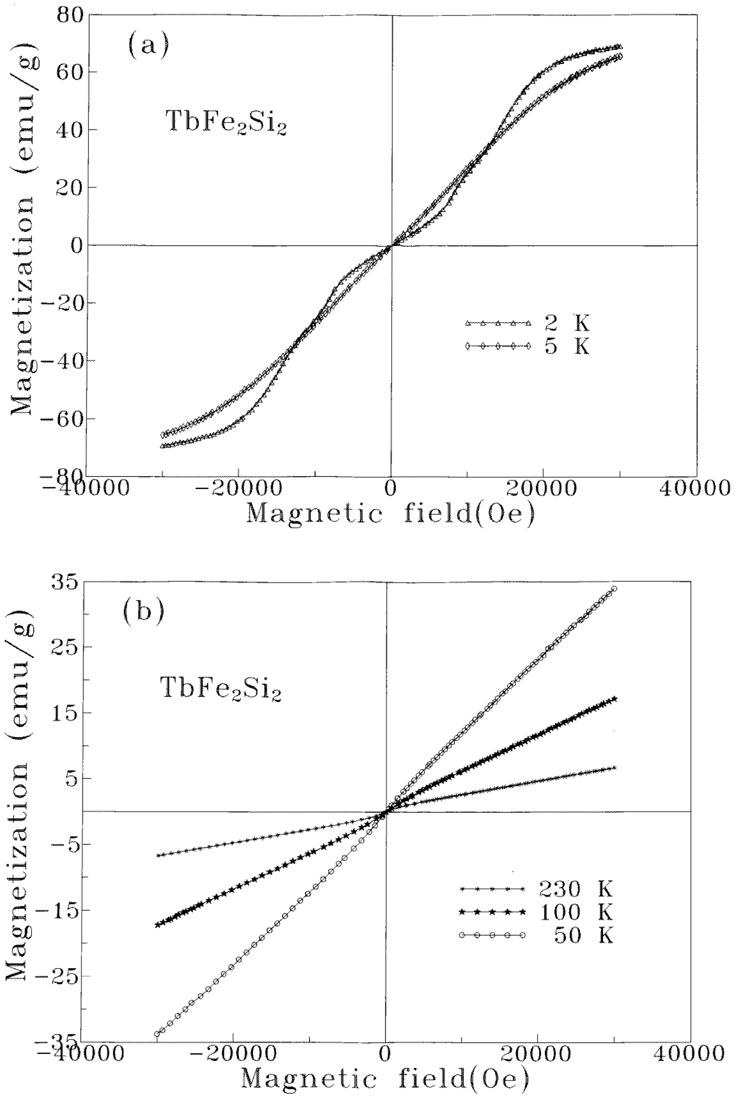

Figure 3. The magnetization curves of $TbFe_2Si_2$: (a) at 2 and 5 K, (b) at 50, 100 and 230 K.

at higher field. Such behaviour is reminiscent of the metamagnetic transitions and implies a spin flip induced by an applied field in an antiferromagnetic system [3]. At $T = 5$ K, the changes in slope are not so pronounced as that at $T = 2$ K. An absence of hysteresis implies that the metamagnetic transitions are second order [4]. We found that the critical fields of the metamagnetic transitions are temperature dependent and decrease as the temperature decreases.

At $T > T_N$, the $M(H)$ curves are composed of two parts (figure 3(b)). The nonlinear part, observed at low applied fields, shows a ferromagnetic-like behaviour, and the linear part at higher fields is typical of a paramagnet. For the temperatures 50, 100 and 230 K, the linear behaviour of the magnetization is achieved at the fields 7, 4.5 and 3 kOe, respectively. With increasing temperature, the slope of the $M(H)$ curves decreases. When we plotted

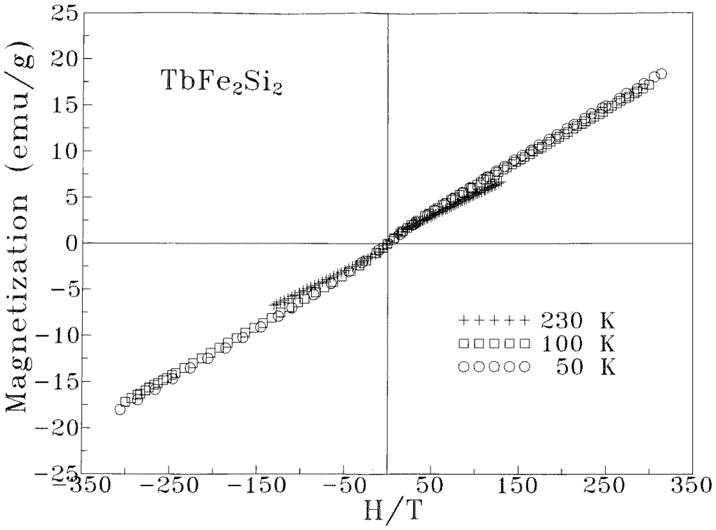

Figure 4. The magnetization as a function of H/T in $TbFe_2Si_2$ at 50, 100 and 230 K.

the magnetization as a function of H/T (figure 4), we found that the two curves for $T = 50$ and 100 K superimpose, and the slope of the $M(H/T)$ curve for $T < T_{SC}$ is higher than that for $T > T_{SC}$. Although the above features of $M(H)$ and $M(H/T)$ are signatures of superparamagnetic behaviour [3], the system may not consist of distinct superparamagnetic particles, and may instead consist of some localized correlated regions which may be dynamic in nature. Thus, at $T_N < T < T_{SC}$, the magnetic susceptibility, as well as the temperature and field dependencies of the magnetization, indicate a spin-fluctuation behaviour in $TbFe_2Si_2$.

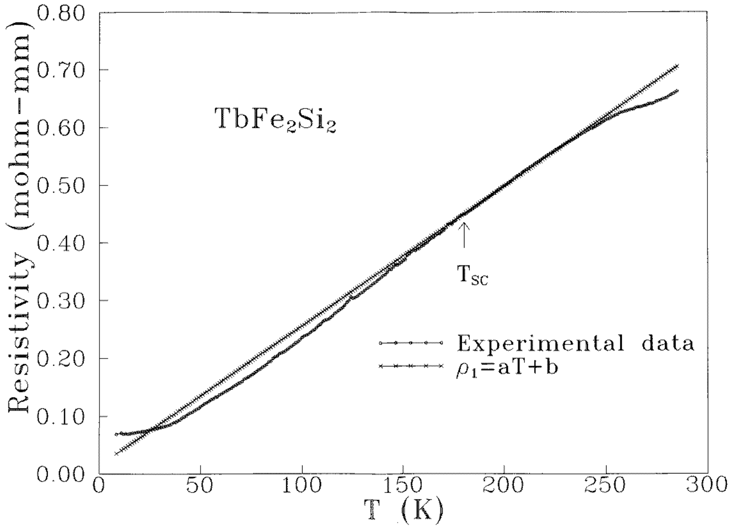

Figure 5. The temperature dependence of the resistivity for $TbFe_2Si_2$.

To obtain more data on these spin fluctuations, we have measured the temperature dependence of resistivity in $TbFe_2Si_2$ (figure 5). At $5 < T < 38$ K, the resistivity curve can be fitted to a temperature-squared law. At higher temperatures, the resistivity is roughly linear over a rather broad range between 70 and 250 K. However, an obvious deviation from linearity is observed at about T_{SC}, indicating a change in transport properties. The experimental data between 180 and 250 K may be fitted well by the $\rho_1(T) = aT + b$ law, and the data between 70 and 180 K may be fitted well by the $\rho_2(T) = cT + d$ law. This indicates that the slope of the $\rho(T)$ curve changes at 180 K. The magnetic anomalies also start to appear at about this temperature (see figures 1 and 2). It can be seen in figure 5 that at $T < T_{SC}$, the resistivity of $TbFe_2Si_2$ deviates downward from the law $\rho_1(T) = aT + b$; i.e. the resistivity is lower at $T < 180$ K. This indicates that, even if some small degree of impurity exists in $TbFe_2Si_2$, the transition at T_{SC} is not due to the impurity. The effect of impurity would be to produce a substantial electron scattering, and one expects an increase of the resistivity below T_{SC}. This implies that the observed anomalies of the resistivity and magnetic behaviour are intrinsic properties of the electronic system of $TbFe_2Si_2$.

4. Discussion

A similar deviation of the magnetic susceptibility from the Curie–Weiss law has been observed in the perovskite systems $RNiO_{3-2\delta}$ (R is a rare-earth element) [5]. The high value of the estimated Pauli susceptibility (such as that for our sample) has been interpreted assuming a correlated gas of ferromagnetically coupled spin polarons [6]. On the other hand, an anomalous behaviour of the susceptibility with a high Pauli term has been observed in the Co-based Laves phases $R(Co_{1-x}M_x)_2$ (M = Al, Fe, Si, Sn, Ga), which have the characteristics of a strongly exchange-enhanced Pauli paramagnet, and possesses the properties of itinerant-electron metamagnetism (i.e. a field-induced transition from the paramagnetic to the ferromagnetic state). The theoretical concept of this phenomenon is based on the spin-fluctuation model [7]. In our $TbFe_2Si_2$, metamagnetic behaviour was observed in the antiferromagnetic state, but its unusual features (figure 3(a)) may perhaps also be related to the itinerant electrons.

T-linear resistivity represents the temperature dependence of the resistivity based on the electron–phonon scattering mechanism. But an alternative mechanism for the origin of T-linear resistivity is the scattering due to spin fluctuations. Recently, a correlation between the deviation of the resistivity from T-linearity and the change in the spectrum of spin fluctuations was found in the high-temperature superconductors YBCO [8–11]. This deviation corresponds to the gap formation in the spin-excitation spectrum (the ‘spin-gap’ effect), as suggested from neutron and NMR studies [12, 13]. It was shown [8, 11] that, if a spin gap exists, $\rho(T)$ and $\chi(T)$ will deviate from linear behaviour before the compound transforms from a paramagnetic to an antiferromagnetic state. In our $TbFe_2Si_2$, spin fluctuations appear at $T < T_{SC}$ before the compound transforms into the antiferromagnetic state. At these temperatures, the charge transport may be influenced by the spin excitations.

Several features of the magnetic behaviour of $TbFe_2Si_2$, observed at $T_N < T < T_{SC}$, may be analogous to those of a superparamagnet, since superparamagnetic behaviour is also explained in terms of a spin-relaxation mechanism [3]. Energy fluctuations can overcome the anisotropy force and spontaneously reverse the magnetization of a superparamagnetic particle from one easy direction to the other. This energy barrier may serve as a gap in the spin excitations.

An unusual decrease of resistivity with temperature has been recently found for the cerium ternary compounds $CeMX_2$ (where M = a transition metal and X = a semimetallic element), and it was attributed to the onset of coherent Kondo lattice ground states [14]. Kondo lattices are characterized by a Fermi-liquid behaviour, which gives rise to a T^2-dependent resistivity. The T^2-dependence of the resistivity found for our sample at low temperatures may also be related to the Kondo lattice effect in $TbFe_2Si_2$.

To verify the mechanism of the spin fluctuations, further experiments, such as SANS or NMR ones, are necessary.

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