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Spin Correlation Effects in $TbFe_2Ge_2$

H. M. Duh, Y. C. Chen, H. F. Chen, C. H. Lin
Department of Physics, National h'aohsiung Normal University,
Kaohsiung, Taiwan 802, R. O. C.

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The detailed studies of magnetic properties and resistivity in the ternary intermetallic compound $TbFe_2Ge_2$ are present. Besides the low temperature antiferromagnetic transition at $T_N = 8.5$ K, the second anomaly in magnetic and electric properties have been found at about $T_{SC} = 250$ K. Below T_N, a metamagnetic behavior 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 resistivity and magnetic susceptibility are related to an effect of spin correlations. Several mechanisms of the spin fluctuations are discussed.

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I. Introduction

The rare earth ternary intermetallic compound $TbFe_2Ge_2$ has the $ThCr_2Si_2$ type crystal structure with space group $I4/mmm$[1]. The dimension of the tetragonal unit cell is approximately 4 × 4 x 10 Å^3. The neutron diffraction study [1] revealed that at low temperatures $TbFe_2Ge_2$ transforms from a paramagnetic state into an antiferromagnetic (AF) state, and the Neel temperature T_N is 7.5 K. Only Tb atoms possess a magnetic moment and the magnetic structure is incommensurate with the crystallographic unit cell. The ^{57}Fe Mossbauer spectroscopy measurements by Noakes et al. [2] have proved that Fe atoms in $TbFe_2Ge_2$ possess no magnetic moments, and the Fe atoms occupy only one type of local sites, i.e., there is no site exchange of the Fe and Ge atoms. A. Szytula etc. [3] solved the magnetic structure of $TbFe_2Ge_2$. It exhibits a modulated antiferromagnetic transition at 8.5 K and a metamagnetic transition with a critical field $H_c = 11$ kOe at low temperature. In this paper we present detailed measurements of resistivity and magnetic properties of the polycrystalline $TbFe_2Ge_2$ compound. Besides the antiferromagnetic transition at 8.5 K, we have found an additional magnetic phase transition at high temperature.

II. Experimental

Polycrystalline $TbFe_2Ge_2$ samples were prepared by melting of stoichiometric amount of constituent materials in an argon arc furnace. The purities of the materials were Tb:3N5, Fe:6N and Ge:6N. To improve homogeneity, the resulting metallic buttons were 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 7 days.

The X-ray measurements were performed with MAC MXP3 X-ray Diffractometer. The X-ray diffraction spectrum of $TbFe_2Ge_2$ can be fitted well to the body center tetragonal crystal structure of the $ThCr_2Si_2$ type with space group $I4/mmm$. The unit cell parameters a and c are 3.9692 Å and 10.273 Å, respectively, and they are consistent with data of Ref. [1].

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

III. Results and analysis

Figure 1 shows the temperature dependence of the inverse susceptibility of $TbFe_2Ge_2$ in the field $H = 2$ kOe. Two anomalies are observed in the $1/\chi$=f(T) dependence. The low temperature anomaly at 8.5 K is typical of a magnetic phase transition from a paramagnetic into an antiferromagnetic state, and our value of T_N = 8.5 K is the same as the result of Szytula et al. [3]. The second anomaly appears at around T_{SC} = 250 K as the 1/X(T) curve deviates from the linear behavior (Fig. 1). It may be an indication of the second phase transition which has not been found in the previous studies.

From a slope of the linear part in 1/X(T) curve (between 250 and 300 K) we calculated the value of magnetic moment μ_{eff} = 12.17 μ_B. Since Fe atoms possess no magnetic moment in $TbFe_2Ge_2$, we have to allow that all moment comes from Tb atoms. However,

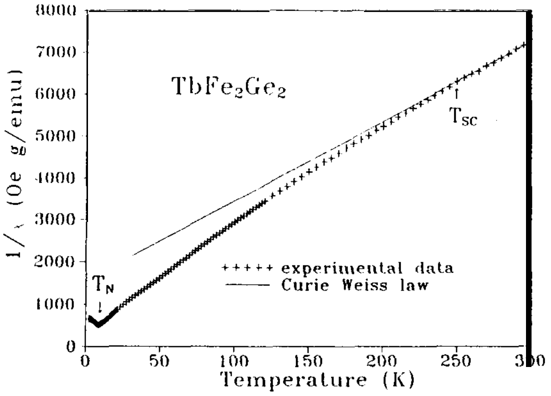


FIG. 1. The temperature dependence of the inverse magnetic susceptibility in $TbFe_2Ge_2$ measured in an applied field of 2 kOe.

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 [I], the value of μ_{eff} was not found for $TbFe_2Ge_2$, but for the similar compounds with the 3d-elements Co, Ni and Cu it is in the range of 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 fit to the law:

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

were χ_0 is the temperature independent contribution due mainly to the itinerant electrons Pauli paramagnetism. When we substrate a constant χ_0 from χ, the change of l/(X $-\chi_0$) is not linear. Therefore the slope l/C will be changed, i.e. the calculated effect magnetic moments will be lowered. The best fit gives the Pauli term of χ_0 in range 3.4 $\sim$ 4.1 x 10^{-5}emu/gOe, and then the Tb magnetic moment reduces to the reasonable value of μ_{eff} (9.2 $\sim 8.6\mu_B$).

It is seen in Fig. 1 that at $T < T_{SC}$, the inverse susceptibility deviates downwards the linear dependence. This implies that below 250 K, an extra spin magnetic moment appears, in addition to the Curie-Weiss and Pauli paramagnetism. If the applied magnetic field is increased to 5 kOe or 10 kOe (not shown in figure), T_{SC} is shifted to low temperature. In order to identify the nature of the extra magnetic moment, the temperature dependence of the magnetization M(T) of $TbFe_2Ge_2$ 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 Fig. 2. We observed that at T_N, the peak positions of the magnetization in two curves are nearly the same. Below T_N, the FC and ZFC curves are split and the FC magnetization is higher than the ZFC one. Above T_N, the ZFC and FC curves superimpose. This behavior indicates that there is no (or very small) spontaneous magnetic moment at $T_N < T < T_{SG}$. Thus the extra magnetic moments may be not a spontaneous magnetic moments.

The field dependencies of the magnetization M(H) of $TbFe_2Ge_2$ at the temperatures below and above T_N are shown in Figs. 3a and 3b, respectively. At all temperatures no

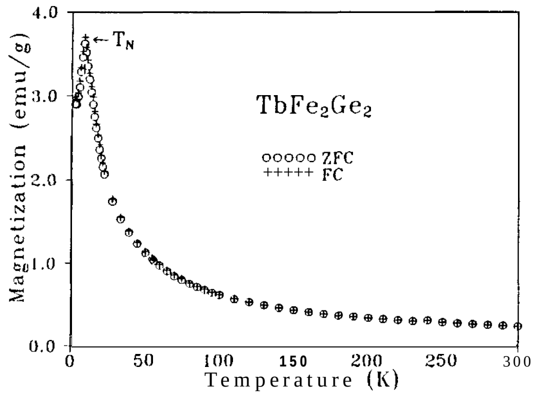
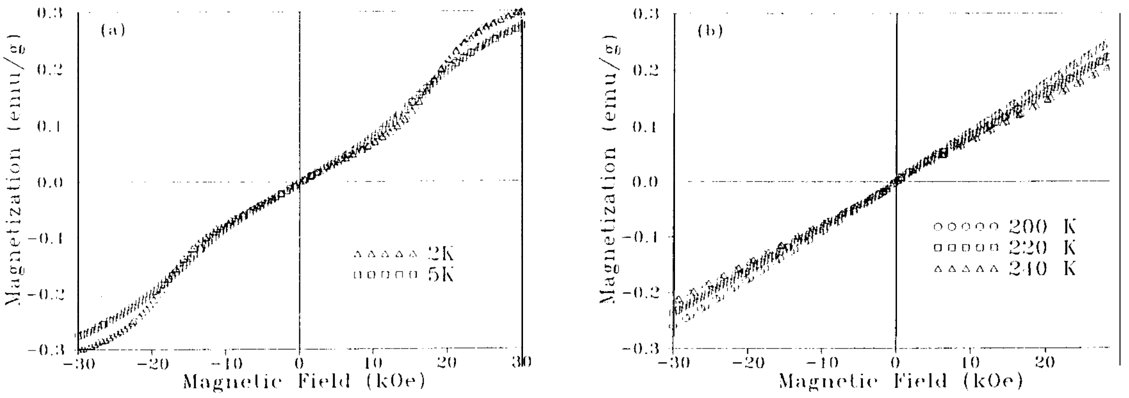


FIG. 2. The temperature dependence of the magnetization in $TbFe_2Ge_2$ for ZFC and FC regimes in the applied field of H = 2 kOe.



(a) (b)

FIG. 3. The magnetization curves of $TbFe_2Ge_2$: (a) at 2 and 5 K, (b) at 200, 220 and 240 K

hysteresis is observed. At T = 2 K, the magnetization curve is rather unusual (Fig. 3a). With increasing the field, the magnetization increases linearly at low field, then it changes the slope several times at about H = 10 and 18 kOe, and then tends to saturate at higher field. Such behavior is reminiscent of the metamagnetic transitions and implies a spin flip induced by an applied field in an antiferromagnetic system [4]. The metamagnetic transition had been observed by Szytula group [3] also. At T = 5 K, the changes in slope are not so pronounced as that at T = 2 K. An absence of hysteresis implies the second-order metamagnetic transitions [5]. 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 compose of two parts (Fig. 3b). The nonlinear part, observed at low applied fields, shows a ferromagnetic - like behavior, and the linear part at higher fields is typical of a paramagnet. With increasing temperature, the slope of M(H) curves decreases. When we plot the magnetization as a function of H/T (Fig. 4), we found that three curves for T = 200, 220 and 240 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 behavior [4], the system may not consist of distinct superparamagnetic particles but may 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 magnetization indicate a spin fluctuation behavior in $TbFe_2Ge_2$.

To get more data about these spin fluctuations, we have measured the temperature dependence of resistivity in $TbFe_2Ge_2$ (Fig. 5). An anomaly appeared at 8.5 K. It corresponds to the antiferromagnetic transition. It can be seen in Fig. 5 that at $T < T_{SC}$, the resistivity of $TbFe_2Ge_2$ deviates downward form the law $[\rho_1(T) = aT + b]$, i.e. the resistivity is lower at T < 250 K. This indicates that, even if some small impurity exists in $TbFe_2Ge_2$, the transition at T_{SC} is not due to the impurity. The effect of impurity would produce a large electron scattering, and one expects an increase of resistivity below T_{SC}. This implies that the observed anomalies of resistivity and magnetic behavior are intrinsic properties of the electronic system of $TbFe_2Ge_2$.

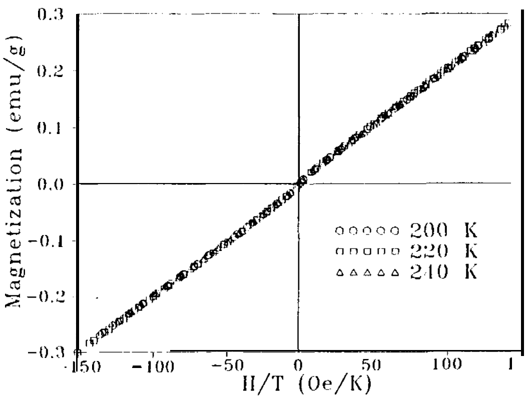


FIG. 4. The magnetization as a function of H/T in $TbFe_2Ge_2$ at 200, 220 and 240 K.

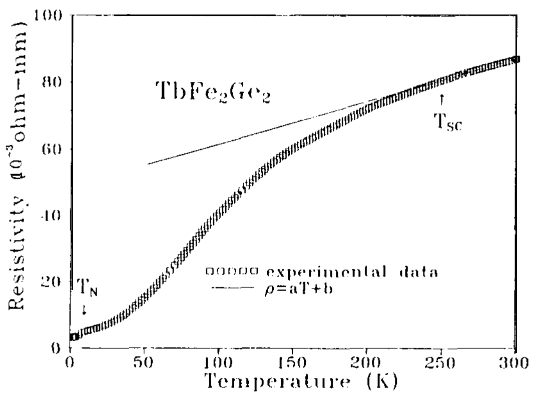


FIG. 5. The temperature dependence of the resistivity for $TbFe_2Ge_2$.

IV. Discussion

The similar deviation of magnetic susceptibility from the Curie-Weiss law have been observed in perovskite systems $RNiO_{3-\delta}$ (R is the rare-earth element) [6]. The high value of the estimated Pauli susceptibility (as that in our sample) has been interpreted assuming a correlated gas of ferromagnetically coupled spin polarons [7]. On the other hand, an anomaly behavior of susceptibility with a high Pauli term have been observed in the Co-based Laves phases $R(Co_{1-x}M_x)_2$, (M = Al, Fe, Si, Sn, Ga), which have characteristics of a strongly exchange-enhanced Pauli paramagnet, and poses the properties of itinerant electron metamagnetism (it is a field-induced transition from the paramagnetic to the ferromagnetic sate). The theoretical concept of this phenomenon is based on the spin fluctuation model [8]. In our $TbFe_2Ge_2$, the metamagnetic behavior was observed in the antiferromagnetic state, but its unusual features (Fig. 3a), perhaps, may also be related to the itinerant electrons.

T-linear resistivity represents the temperature dependence of 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 high temperature superconductors YBCO [9-12]. This deviation corresponds to the gap formation in the spin excitation spectrum (the "spin gap" effect), as suggested from neutron and NMR studies [13,14]. It was shown [9,12] that, if a spin gap exists, p(T) and X(T) will deviate from linear behavior before the compound transforms from a paramagnetic to an antiferromagnetic state. In our $TbFe_2Ge_2$, 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 magnetic behavior of $TbFe_2Ge_2$, observed at $T_N < T < T_{SC}$, are typical of a superparamagnet. The superparamagnetic behavior is also explained in terms of a spin relaxation mechanism [4]. 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.

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

Acknowledgments

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