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Transport properties of the itinerant-electron weak ferromagnet $LaFe_4As_{12}$

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ABSTRACT

We have investigated the electrical resistivity under magnetic fields in the itinerant-electron weak ferromagnet $LaFe_4As_{12}$, in order to clarify the characteristics of spin fluctuations in this material by comparison with the reported typical itinerant-electron ferromagnets. As a common feature to the typical itinerant-electron weak ferromagnets, strong enhancement of negative longitudinal magnetoresistivity was observed near T_C, reflecting the suppression of spin fluctuations. As a unique behavior of $LaFe_4As_{12}$, we found a considerable positive component of the magnetoresistivity above T_C.

1. Introduction

The filled skutterudite compounds with a chemical formula MT_4X_{12} (M = alkaline, alkaline-earth, rare-earth, or actinide metal; T = Fe, Ru, or Os; and X = P, As, or Sb) crystallize in the cubic $LaFe_4P_{12}$ structure [1]. These compounds have been intensively investigated, since they show various attractive features such as heavy fermion superconductivity, M–I transition, etc. [2–4]. However, on the filled skutterudite arsenides (MT_4As_{12}), only limited investigations have been reported because of the difficulty in synthesizing high-quality samples. Recently, three ferromagnets M = La, Pr, and Sm have been found in MFe_4As_{12} [5–7]. $PrFe_4As_{12}$ and $SmFe_4As_{12}$ were reported to exhibit a ferromagnetic transition at 18 and 39 K, respectively. As a common feature to $PrFe_4As_{12}$ and $SmFe_4As_{12}$, the magnetic measurements suggest that both the itinerant 3d electrons and localized 4f-electrons contribute to the magnetic state in these compounds. $LaFe_4As_{12}$ is the first La-based filled skutterudite exhibiting itinerant-electron weak ferromagnetism below $T_C = 5.2$ K with no 4f-electron, and is the promising material close to a ferromagnetic quantum critical point (QCP).

As another view point, for $LaFe_4X_{12}$ (X = P, As, Sb), the electronic density of states at Fermi level DOS(E_F) was predicted to vary systematically with X, according to the band structure calculation [8]. $LaFe_4P_{12}$ with the lowest DOS(E_F) shows Pauli paramagnetic behaviors just like an ordinary metal [9], while $LaFe_4Sb_{12}$ with the highest DOS(E_F) is an enhanced Pauli paramagnet close to a ferromagnetic QCP exhibiting non-Fermi liquid behaviors [10]. $LaFe_4As_{12}$ with the medium DOS(E_F) was found to exhibit itinerant-electron weak ferromagnetism. The ferromagnetism is unexpected within the simple Stoner-type argument. This fact brings about new question why the ferromagnetism is stabilized in the system with medium DOS(E_F). There may be a clue to solve the problem in the difference of temperature dependence of electrical resistivity ρ at low temperatures. ρ for $LaFe_4P_{12}$ at low temperatures exhibits T^2-dependence expected for the Fermi liquid, while ρ for both $LaFe_4As_{12}$ and $LaFe_4Sb_{12}$ exhibits $T^{5/3}$-dependence. The $T^{5/3}$-dependence suggests an evident role of the critical fluctuation of the three-dimensional weak ferromagnet in the latter two compounds [6,10]. We found that the coefficient of the $T^{5/3}$-dependence in $LaFe_4Sb_{12}$ is several times larger than that in $LaFe_4As_{12}$. Based on these facts, one can infer that the larger spin fluctuations in $LaFe_4Sb_{12}$ prevent the ferromagnetic transition in this material.

In our previous study, we reported the negative transverse magnetoresistivity (MR) in $LaFe_4As_{12}$ at 4.2 K. However, the transverse MR is not suited for extracting the spin fluctuation contribution, since the ordinary positive MR due to the cyclotron motion of conduction electrons is larger. To further investigate the effect of spin fluctuation scattering, the temperature and magnetic field dependence of the electrical resistivity has been systematically measured on a $LaFe_4As_{12}$ single crystal.

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2. Experimental

We have succeeded in growing high-quality single crystal of $LaFe_4As_{12}$ under high-pressure ~4 GPa. The starting elements, chips of La (99.9%) and powders of Fe (99.99%) and As (99.9999%) were mixed in the ratio of 1 : 4 : 20. Those elements were placed in a cylindrical BN crucible and compressed to ~4 GPa by using a cubic-anvil high-pressure apparatus. They were heated up to 1200 K, then cooled to 1100 K at a rate of 4 K/h and quenched down to room temperature. The details about sample preparation were reported in Ref. [11].

X-ray powder diffraction (XRD) measurements were performed on the obtained single crystals of $LaFe_4As_{12}$. All the diffraction peaks are indexable as those of the filled skutterudite structure without any impurity phase, in contrast with a small amount of impurity phases in the polycrystalline samples in our previous report. The electrical resistivity under magnetic fields was measured with the ordinary four probe method by using a Quantum Design physical property measurement system down to 2 K and 9 T.

3. Results and discussion

Fig. 1 shows the temperature dependence of the electrical resistivity $\rho(T)$ in $LaFe_4As_{12}$. At around 5.2 K, $\rho(T)$ shows an evident kink reflecting the ferromagnetic transition. The residual resistivity ratio (RRR) is ~147, which is larger than twice that of the polycrystalline sample in the previous report, reflecting the reduced impurity phases. In fact, we have recently succeeded in observing clear de Haas–van Alphen oscillations on a sample grown by the same procedure, which indirectly evidences the high quality of the present sample [12].

Fig. 2 shows the field dependence of the longitudinal magnetoresistivity (LMR) and transverse magnetoresistivity (TMR) $\Delta\rho/\rho(0) = \{\rho(H) - \rho(0)\}/\rho(0)$ at 4 K (upper panel) and 12 K (lower panel). In the upper panel, the strong enhancement of negative MR is observed at 4 K. According to the self-consistent renormalization (SCR) theory [13], as a result of the reduced spin fluctuation scattering, the negative MR is proportional to H in the ferromagnetic state under low magnetic fields. The total $\Delta\rho/\rho(0)$ can be described as a combination of spin fluctuations term and the H^2-dependent contribution from the ordinary MR due to the cyclotron motion of the conduction electrons as follows:

$$\Delta\rho/\rho(0) = -aH + bH^2, \tag{1}$$

where a and b are positive constants. The experimental data of LMR and TMR roughly obey Eq. (1) indicated by the solid curves in the upper panel. Negative TMR is observed only in low fields, while for LMR the spin fluctuation term is dominant over a wide range of magnetic field. That is naturally understood, since the contribution of the cyclotron motion to LMR is smaller. On the other hand, in the paramagnetic state at 12 K, both TMR and LMR are positive as shown in the lower panel. According to the SCR theory, the negative MR due to the suppression of spin

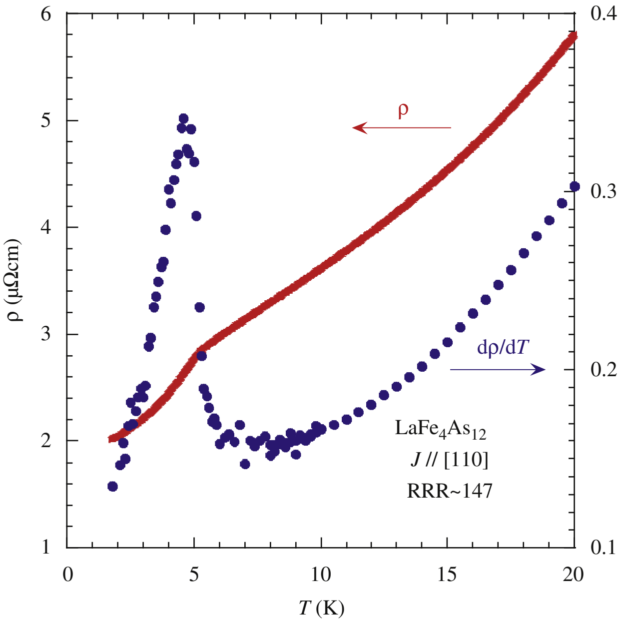

Fig. 1. Electrical resistivity ρ and the temperature derivative of electrical resistivity $d\rho/dT$.

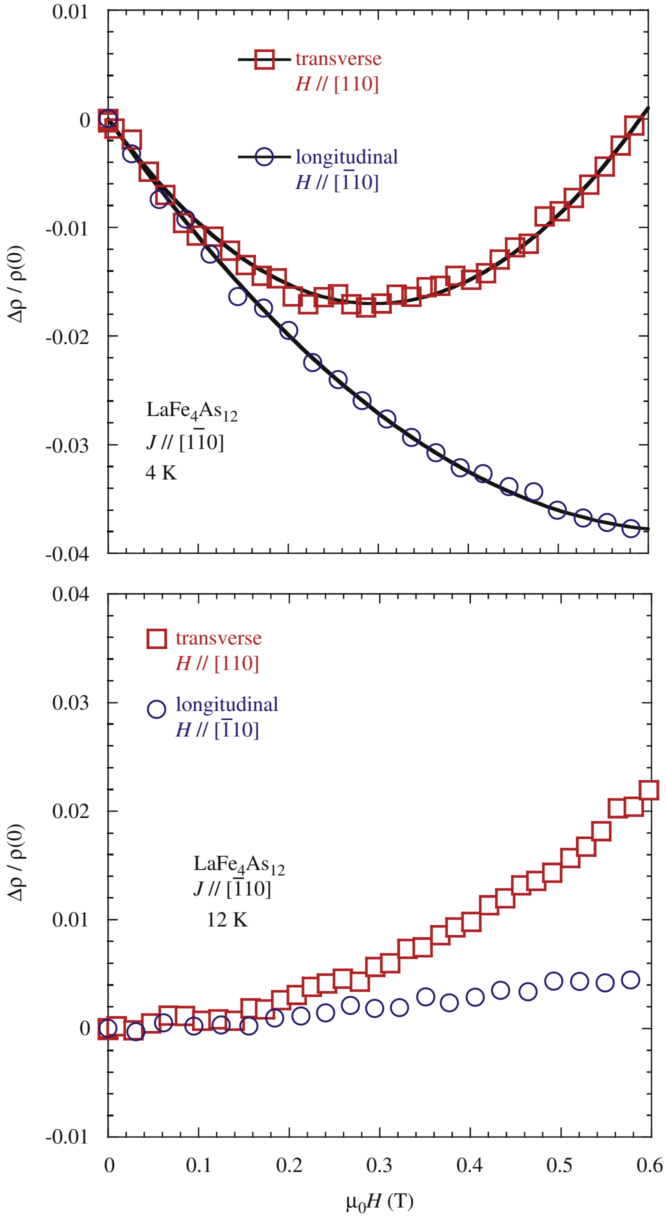

Fig. 2. Comparison of the field-dependent parts of the longitudinal and transverse magnetoresistivity $\Delta\rho/\rho(0) = \{\rho(H) - \rho(0)\}/\rho(0)$. The solid curves in the upper panel represents a fit of Eq. (1) to the experimental data.