Magnetic Properties of Itinerant Ferromagnet La$_{12}$Fe$_{57.5}$As$_{41}$

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Abstract. We successfully synthesized the single crystal of the itinerant ferromagnetic compound La$_{12}$Fe$_{57.5}$As$_{41}$ by Sn-flux method and studied its magnetic properties by magnetization measurements. The temperature dependence of magnetic susceptibility obeys the modified Curie-Weiss law, $\chi^{-1} = (\chi_0 + C/(T - \theta))^{-1}$, quite well in the high temperature region. The effective moment $P_{eff}$ and the Weiss temperature $\theta$ are estimated to be 1.13 $\mu_B$/Fe and 128.5 K, respectively. The magnetization of La$_{12}$Fe$_{57.5}$As$_{41}$ shows the convex behavior when plotted in a form of $M^2$ vs $H/M$ at low temperatures, while the $M^4$ shows the linear relation against $H/M$ in a wide temperature range near $T_C$.

1. Introduction
La$_{12}$Fe$_{57.5}$As$_{41}$ is a newly discovered itinerant ferromagnet with the Curie temperature $T_C = 125$ K$[1]$. It has the orthorhombic crystal structure with the space group of $Pnmn$. The complex three-dimensional network of La$_{12}$Fe$_{57.5}$As$_{41}$ is composed of novel wave-like layers of connected As-centered trigonal prisms$[1]$. The Fe atoms form the wave-like square lattice, which is parallel to the $ab$ plane$[1]$. It is quite different with the FeAs layers in the mother compounds of Fe-based superconductors and the three-dimensional network in the CaFe$_4$As$_3$$[2]$, both of which show the antiferromagnetic order at low temperatures. Therefore, it is very interesting to investigate the magnetic properties of La$_{12}$Fe$_{57.5}$As$_{41}$ due to the rare examples of ferromagnetic order in Fe-As contained compounds.

However, the ferromagnetic impurities are often introduced into the polycrystalline samples, which mask the intrinsic magnetic properties of La$_{12}$Fe$_{57.5}$As$_{41}$$[1]$. The high quality samples are strongly needed to elucidate the magnetic properties of this compound. In this work, we successfully synthesized the single crystal samples of La$_{12}$Fe$_{57.5}$As$_{41}$ and investigated its magnetic properties by the measurements of magnetic susceptibilities and isothermal magnetizations with the applied magnetic field parallel to the $ac$ plane.

2. Experiments
The single crystal of La$_{12}$Fe$_{57.5}$As$_{41}$ was synthesized by the Sn-flux method. A 2 g mixture of La (Lump 99.9%), Fe (bulk 99.99%), and As powder (99.999%) in a molar ratio of 1:5:3 was
placed in an evacuated silica tube together with 12 g of Sn (99.99%). The mixture was heated to 1050°C over 12 h, held at this temperature for 12 h, and then slowly cooled to 600°C with 3°C/h, followed by furnace cooling to room temperature. Black needle-shaped crystals of the ternary arsenides La$_{12}$Fe$_{57.5}$As$_{41}$, together with crystals of binary-phase byproduct FeAs, were gained after dissolving excess Sn flux in diluted acid HCl. The magnetization was measured using a superconducting quantum interference device magnetometer in the temperature range between 2 and 300 K with an applied magnetic field $H$ up to 5 T.

3. Results and Discussion

Figure 1 shows the temperature dependence of the magnetization divided by the external magnetic field $M/H$ (■) and the inverse magnetic susceptibility (□) for La$_{12}$Fe$_{57.5}$As$_{41}$ measured under $H = 0.5$ T with the magnetic field parallel to the $ac$ plane. The ferromagnetic phase transition manifest itself by a rapid increase of the magnetic susceptibility at about 125 K with further decreasing the temperature, which is similar to the result of reference 1. However, the absolute magnitude of the susceptibility at low temperature is only about 25% of the value of susceptibility reported previously[1]. As we will point out below, this indicates that our sample is very pure without any magnetic impurities. On the other hand, the magnetic susceptibility obeys the modified Curie-Weiss law, $\chi^{-1} = (\chi_0 + C/(T - \theta))^{-1}$, quite well in high temperature region (150 K ~ 300 K) as shown in Fig. 1 by solid line. As a result, the temperature independent part $\chi_0$, Curie constant $C$ and Weiss temperature $\theta$ are estimated to be 0.016 emu/mol, 9.14 emu K/mol and 128.5 K, respectively. From the Curie constant $C$ we obtained the effective moment $P_{eff} = 1.13 \mu_B$/Fe.

Figure 1. Temperature dependence of the magnetization divided by the external magnetic field $M/H$ (■) and the inverse magnetic susceptibility (□) for La$_{12}$Fe$_{57.5}$As$_{41}$ with the external magnetic field applied parallel to the $ac$ plane.

Figure 2 shows the isothermal magnetization curves of La$_{12}$Fe$_{57.5}$As$_{41}$ from 2 K to 190 K under the external magnetic field up to 5 T. The magnetization saturates in the field of 2 T at 2 K. Moreover, with the increase of temperature, the saturated magnetization decreases continuously in the ferromagnetic ordered state. The linear relationship between $M$ and $H$
can be found at the temperature higher than 170 K. Our results are quite different with that reported by S. S. Stoyko et al., where the linear relation is absent even in the paramagnetic state with the temperature up to 300 K[1]. This is because of the existence of the magnetic impurities in their sample as they claimed. These results suggest our sample is of high quality.

![Figure 3. Arrott Plot (M^2 versus H/M plot) for La_{12}Fe_{57.5}As_{41} at various temperatures.](image)

![Figure 4. M^4 versus H/M plot for La_{12}Fe_{57.5}As_{41} at various temperatures.](image)

On the basis of the Stoner model, the high field isothermal magnetization follows $M^2 = A + B(H/M)$, where the parameters $A$ and $B$ are independent of magnetic field $H$. This leads to a series of linear and parallel curves in $M^2$ versus $H/M$ plots that are commonly so-called Arrott plots. This behavior has been observed in some magnetic materials, such as ZrZn$_2$[8], Y(Co-Al)$_2$[4, 5], Ni$_3$Al[6], Sc$_3$In[7]. Figure 3 shows the $M^2$ vs $H/M$ plots at various temperatures from 2 K to 150 K. The linear relationship between $M^2$ and $H/M$ can be found with the temperature lower than 80 K. Usually, the Curie temperature $T_C$ can be determined at the temperature, where the intercept along the $M^2$ axis in the Arrott plot crosses the origin of the coordinate axes, whereas, in the temperature range between 90 K and 130 K, the $M^2$ does not show the linear relation but a convex curvature against $H/M$. It is difficult to determine the $T_C$ from the Arrott plot in this case. However, if we plot them in a $M^4$ vs $H/M$ form, it is clear that $M^4$ shows good linear relation with $H/M$ in a wide temperature range near $T_C$ in La$_{12}$Fe$_{57.5}$As$_{41}$ as shown in Fig. 4. This behavior has been observed in some weak itinerant ferromagnetic materials, such as MnSi[8], LaCoAsO[9]. The linear relationship of $M^4$ with $H/M$ is explained on Takahashi’s theory of spin fluctuations, which assumed that the sum of zero-point and thermal spin fluctuations is independent of temperature[10]. Therefore, $T_C$ is estimated to be 125 K approximately from the $M^4$ vs $H/M$ plot.

In order to clarify a magnetic material belongs to the itinerant or a localized spin system, we estimate the Rhodes-Wohlfarth ratio of La$_{12}$Fe$_{57.5}$As$_{41}$, which is defined as the value of $P_c/P_s$, where $P_c(P_c + 2) = P_{\text{eff}}^2$ and $P_s$ is the spontaneous magnetization at ground state[11]. In the case of the weak itinerant ferromagnet, Rhodes-Wohlfarth ratio is known to be much larger than the case of the localized spin system $P_c/P_s \equiv 1$. We estimate $P_c = 0.51\,\mu_B$/Fe by using the value $P_{\text{eff}} = 1.13\,\mu_B$/Fe. $P_s = 0.17\,\mu_B$/Fe is estimated from the Arrott plot as shown in Fig. 3. Using these values we obtained $P_c/P_s$ of La$_{12}$Fe$_{57.5}$As$_{41}$ is about 3, larger than the 1, which indicates this material belongs to the itinerant ferromagnet class.
4. Summary
The itinerant ferromagnetic compound La$_{12}$Fe$_{57.5}$As$_{41}$ was synthesized successfully by Sn-flux method. The magnetic properties were studied by the measurements of magnetic susceptibility and magnetization with the external magnetic field applied in the ac plane. The temperature dependence of magnetic susceptibility obeys the modified Curie-Weiss law, $\chi^{-1} = (\chi_0 + C/(T - \theta))^{-1}$ quite well in high temperature region. The effective moment $P_{\text{eff}}$ and Weiss temperature $\theta$ are estimated to be 1.13 $\mu_B$/Fe and 128.5 K, respectively. We showed that the magnetization of La$_{12}$Fe$_{57.5}$As$_{41}$ has the linear behavior when plotted in the form of $M^2$ vs $H/M$ in the lower and the higher temperature regions, while changes to the $M^4$ vs $H/M$ in a wide temperature range near $T_C$. The Rhodes-Wohlfarth ratio is estimated to be about 3, suggesting that La$_{12}$Fe$_{57.5}$As$_{41}$ is an itinerant ferromagnet.

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