

Anomalous magnetization in the layered itinerant ferromagnet LaCoAsO

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We synthesized weakly itinerant ferromagnet with layered structure LaCoAsO and measured the isothermal magnetizations at various temperatures. The isothermal magnetizations in a form of Arrott plot for weakly itinerant ferromagnets such as $ZrZn_2$, Sc_3In , Ni_3Al , and $Y(Co, Al)_2$ shows parallel linear relation over the wide temperature range near the Curie temperature T_C , while for LaCoAsO the square of the magnetization M^2 was found to show a convex curvature against H/M , similar to MnSi and $Fe_xCo_{1-x}Si$. We analyzed the magnetization of LaCoAsO at T_C and estimated several parameters of spin fluctuations on the basis of self-consistent renormalization theory of spin fluctuations. We simulated a reciprocal magnetic susceptibility above T_C and discussed the nature of spin fluctuations in LaCoAsO.

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I. INTRODUCTION

Recent discovery of high- T_c iron pnictides^{1,2} has aroused so much interest that many intensive experimental and theoretical studies have been devoted so far to clarifying the superconducting mechanism and to discoveries of related new materials. After the discovery of superconducting $LaFeAs[O_{1-x}F_x]$, several associated iron arsenides, e.g., $LnFeAs[O_{1-x}F_x]$, $LnFeAsO_{1-x}$, $[A_xB_{1-x}]Fe_2As_2$, Li_xFeAs and iron selenide $FeSe_x$, were found to become superconductors, where Ln , A , and B are lanthanoid, alkaline, and alkaline earth metal ions, respectively. For the sake of those efforts, the important features of the superconductors in those iron based systems were clarified: the layered structure with FeAs (or FeSe) planes where Fe ions form two-dimensional (2D) square lattice, and the valence of Fe ions of undoped compounds is +2 when the ionic model is assumed.

The metal iron site of $LaFePnO$ ($Pn=P, As$) can be substituted by other transition metals. Chemical studies about $LnMPnO$ (Ln : lanthanoids, M : transition metals) were reported at first by German groups.³ In the case of manganese, $LaMnPO$ was reported to be an antiferromagnetic semiconductor.⁴ In the case of nickel, $LaNiPO$ was reported to be a superconductor with superconducting transition temperature $T_c \sim 2$ K.⁵ Furthermore, in the case of cobalt, i.e., $LaCoAsO$ and $LaCoPO$ were reported to be ferromagnetic with the Curie temperature T_C of about 50 and 60 K, respectively.^{6,7} $LaCoPnO$ has the tetragonal crystal structure with the space group of $P4/nmm$ (ZrCuSiAs type), which is the same as that of $LaFeAsO$. Cobalt ions form two-dimensional square lattices with sandwiched by pnictide layers. Thus, $LaCoPnO$ is expected to be a ferromagnet with a highly two-dimensional anisotropic magnetic state, which should be related to the high- T_c iron pnictides.

The theoretical study of the itinerant ferromagnets traces back to the Stoner model.⁸ Although this model succeeded in showing the nature of the ordering state, the consequences of this theory such as quite high Curie temperature T_C and non-Curie-Weiss behavior of magnetic susceptibility above T_C did not match with many experiments, remaining unsolved problem for many decades because the Stoner theory was

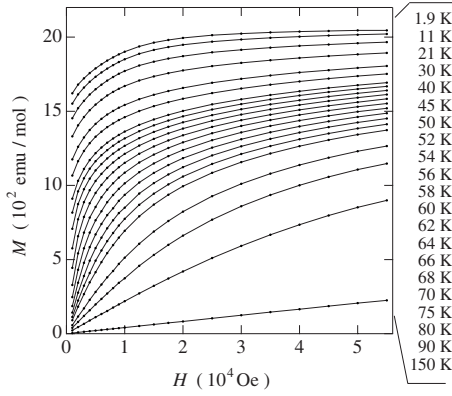
based upon the Hartree-Fock mean-field approximation and almost neglected the effects of spin fluctuations. Moriya and his co-workers self-consistently fed back the effects of spin fluctuations to the magnetic susceptibility, and succeeded in theoretically reproducing the Curie-Weiss behavior of ferromagnetic metals originated in the linear growing of spin fluctuations, e.g., square of local spin density S_L^2 , with T .⁹⁻¹² This self-consistent renormalization (SCR) theory of spin fluctuations succeeded in clarifying the nature of nearly and weakly itinerant (anti-)ferromagnetic metals. Furthermore, Takahashi, one of Moriya's co-workers, developed the SCR theory by assuming that the sum of zero-point and thermal spin fluctuations is conserved against temperature (T).¹³

In this report, we show the magnetic properties of polycrystalline sample of LaCoAsO, which is a weak itinerant ferromagnetic (WIF) compound with two-dimensional layered crystal structure, and report the results of analyses based upon the SCR theory of spin fluctuations. Since we can estimate only one spin-fluctuation parameter from static magnetic measurements, both Takahashi's and the SCR theories are adopted for the analyses of experimental results.

II. EXPERIMENTS

For the synthesis of polycrystalline samples of LaCoAsO, we used powders of La (purity: 99.9%), As (99.99%) and CoO (99.99%) as starting materials. At first, powders of La and As were mixed and put in an evacuated silica tubes. The mixtures of La and As were carefully fired in a furnace at 550 °C for 5 h and then at 800 °C for 12 h. The obtained powders of LaAs were mixed with the powders of CoO to a stoichiometric ratio and ground well in hexane to avoid an oxidation. The pelletized mixtures of LaAs and CoO were put in an evacuated silica tube and fired at 1000 °C for 12 h. Of course the single crystal is more suitable for the study of the anisotropy of magnetization. We have tried to synthesize the single crystal of this compound by tin flux method which was used for the synthesis of $LaCoPO$, we have not obtained the single crystal samples for now. Thus, we synthesized the polycrystalline but high purity sample in this work.

We measured powder x-ray diffractions of the obtained polycrystalline samples and confirmed them to be in a single

FIG. 1. M versus H for LaCoAsO at various temperatures.

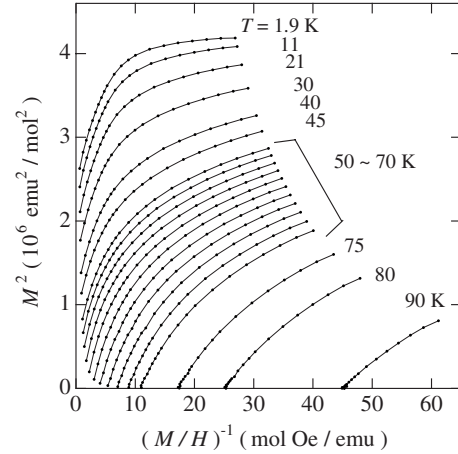
phase of LaCoAsO. The estimated lattice parameters a and c were 4.055 and 8.462 Å, respectively, being in good agreement with the previous reports.^{6,14} The magnetizations (M) of LaCoAsO were measured as functions of T and magnetic field (H) by using MPMS (Quntam Design Inc.). The T dependence of M was almost the same as that of Ref. 6. The M vs H curves were measured by decreasing H from 5.5 T to zero, and each curve was measured by increasing T from 1.9 K after field cooling from room temperature.

III. RESULTS AND DISCUSSION

Figure 1 shows the isothermal magnetization (M) curves in the T range from 1.9 to 250 K. At low temperatures the M showed saturating behavior above 3 T. With increasing T , the saturated magnetization is reduced, and the M comes to be proportional to H above 90 K.

Usually, the T_C is determined from $M(T, H)^2$ vs $H/M(T, H)$ plots, i.e., so called Arrott plots.¹⁵ Generally, the free energy can be expanded by the order parameter around the transition temperature T_C , which is so called Landau expansion, and in the ferromagnets the order parameter is the spontaneous magnetization $M_s(T)$ [$\equiv M(T, 0)$]. If the sixth and higher terms of the expanded free energy are neglected, the relation between H and $M(T, H)$ is given as $H = a \cdot M(T, H) + b \cdot M(T, H)^3$ from the equilibrium equation by using the Landau expansion of free energy under magnetic field. Therefore, in a homogeneous itinerant ferromagnet, $M(T, H)$ obeys the relation of $M(T, H)^2 = M_s(T)^2 + \zeta \cdot H/M(T, H)$, where ζ is the coefficient being independent of temperature around T_C . Therefore, the T_C can be determined by the Arrott plots as the temperature at which a linear line passes the origin because of $M_s(T)$ being zero at T_C .

Figure 2 shows the M^2 vs H/M plots at various temperatures from 1.9 to 90 K. The M^2 does not show the linear relation but does a convex curvature against H/M for every temperature. The Arrott plot of LaCoAsO was reported in Ref. 7. Although their figure showed only the low field region, their result seems quite similar to ours. The convex curvature of M^2 against H/M reminds us of the magnetization of MnSi (Ref. 16) and $\text{Fe}_x\text{Co}_{1-x}\text{Si}$ (Ref. 17) which are typical weakly itinerant ferromagnets. To our knowledge, the

FIG. 2. M^2 versus H/M (Arrott plot) for LaCoAsO at various temperatures.

convex curvature of M^2 against H/M has been reproduced only by Takahashi's developed spin-fluctuation theory so far.¹³ In this theory, it was assumed that the sum of zero-point and thermal spin fluctuations is conserved against T , and as the consequence of this assumption he argued the importance of the sixth coefficient of the free energy when the system has a relatively larger $\eta [= (T_C/T_0)^{1/3}]$ value. Here T_0 characterizes the energy width of the dynamical spin-fluctuation spectrum. In such case, the magnetization obeys following relation at T_C because the fourth coefficient becomes zero:

$$h = [T_A/3(2 + \sqrt{5})T_C]^2 p^5, \quad (1)$$

where $h/2\mu_B$ and p are the magnetic field and the magnetization in μ_B unit, respectively. The parameter T_A characterizes the dispersion of the static magnetic susceptibility in wave vector (q) space. Equation (1) shows that the M^4 obeys linear relation against H/M at T_C and, therefore, T_A can be estimated here without dynamical experiments such as the neutron scatterings and NMR relaxations.

We replotted the isothermal magnetization curves in a form of M^4 vs H/M as shown in Fig. 3. Here, the M^4 obeys almost the linear relation against H/M around $T_C \sim 55$ K. Such a linear relation was also confirmed in MnSi as well as $\text{Fe}_x\text{Co}_{1-x}\text{Si}$.^{13,17} Strictly speaking, M^4 is not linear completely to H/M just around T_C as seen in Figs. 3 and 4(b). However, most of itinerant ferromagnets show the concave behavior when plotted in the form of M^4 versus H/M [see the data of Ni_3Al in Fig. 4(b) (Ref. 18)]. Furthermore, M^4 of the typical itinerant ferromagnetic compound MnSi is also not exactly linear to H/M . According to Takahashi's theory, in the case of the coefficient of M^4 in the Landau expansion of free energy being zero, M^4 becomes linear against H/M . Thus a complete linearity is just obtained in a delicate condition. LaCoAsO is not just on this condition, but seems to be very near the condition as well as the quantum critical point due to spin fluctuations. We show the results of linear fitting to the M^2 and M^4 plotted against H/M in Fig. 4. This figure clearly shows that at $T_C = 55$ K the M^4 shows a higher linearity against H/M than the M^2 against H/M , while Ni_3Al

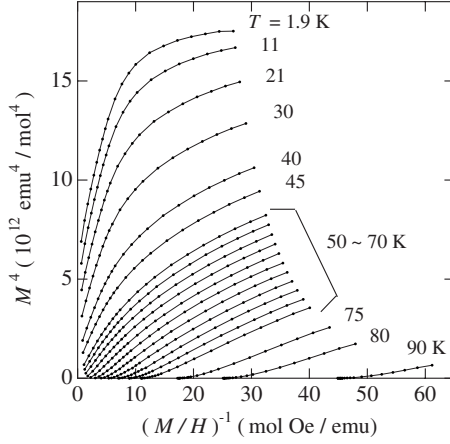


FIG. 3. M^4 versus H/M for LaCoAsO at various temperatures. Around T_C , the M^4 almost obeys the linear relation against H/M .

shows almost complete linearity in M^2 against H/M . Equation (1) can be transformed as the relation of M^4 vs H/M as

$$M^4 = 1.17 \times 10^{19} (T_C^2/T_A^3)(H/M), \quad (2)$$

where M and H are in emu/mol and Oe units, respectively. By putting the value of the slope at $T_C=55$ K into Eq. (2), we obtained T_A as 6.21×10^3 K.

According to the SCR theory for WIF,^{9,11,12} T_C is related with P_s , T_A and T_0 as a following relation:

$$T_C = (60c)^{-3/4} P_s^{3/2} T_A^{3/4} T_0^{1/4}, \quad (3)$$

where c and P_s are a constant equal to $0.3353 \dots$ and the spontaneous magnetization in the ground state $M_s(0)$ in μ_B unit, respectively. The estimation of P_s is described in a following paragraph. By putting the value of T_A determined by Takahashi's theory into Eq. (3), we obtained T_0 as 644 K.

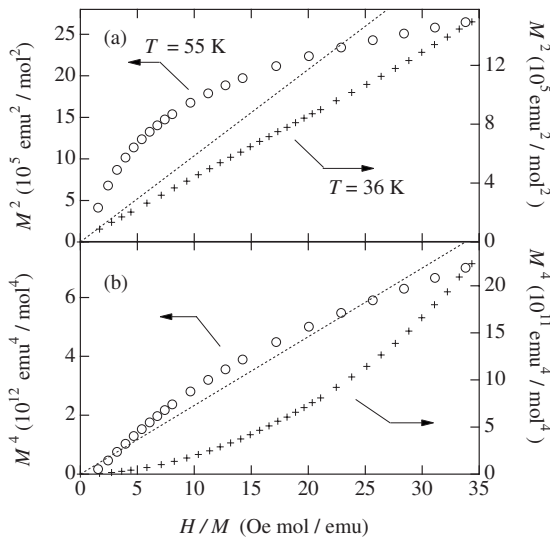


FIG. 4. Linear fitting for (a) M^2 versus H/M and (b) M^4 versus H/M for LaCoAsO at $T=55$ K. Dashed lines are the results of linear fitting to the data. Crosses in both panels are the data of Ni_3Al at $T_C=36$ K.¹⁸

TABLE I. Spin fluctuation parameters of LaCoAsO. The values of \bar{F}_1 denoted (i) and (ii) were estimated from the slope of Arrott plot and from Eq. (5), respectively.

T_C (K)	T_A (10^3 K)	T_0 (10^2 K)	\bar{F}_1 (10^3 K)
55	6.21	6.44	(i) 6.10 (ii) 16.0

In the SCR theory, the coefficient of the M^4 term of the Landau expansion of free energy is one of the most important spin-fluctuation parameters, called \bar{F}_1 , which is usually determined in Kelvin unit as

$$\bar{F}_1 = N_A^3 (2\mu_B)^4 / \zeta k_B, \quad (4)$$

where N_A and k_B are Avogadro's number and the Boltzmann constant, respectively, and ζ is the slope of Arrott plot at T_C . Therefore, for WIF, \bar{F}_1 can be determined uniquely from the slope of the Arrott plot at T_C . In the present case, however, the Arrott plot does not obey linear relation, so we estimated the value of \bar{F}_1 by two ways. We estimated \bar{F}_1 from the slope of a high field region of the M^2 vs H/M curve at T_C . From this way we obtained \bar{F}_1 as 6.10×10^3 K. We also estimated \bar{F}_1 from the following relation:

$$\bar{F}_1 = 4T_A^2/15T_0. \quad (5)$$

This relation was predicted in Takahashi's theory.¹³ By putting the obtained values of T_A and T_0 to Eq. (5), the value of \bar{F}_1 becomes 1.60×10^4 K. The main spin-fluctuation parameters are listed in Table I.

We estimated $M_s(T)$ below T_C and the reciprocal magnetic susceptibility $1/\chi (= \lim_{H \rightarrow 0} H/M)$ above T_C from Figs. 2 and 3, respectively. The M_s and $1/\chi$ are estimated as the values of the intersections of natural extrapolations of the M^2 (M^4) curves with the vertical and horizontal axes, respectively. The values estimated from the M^2 and M^4 curves were almost the same, so we only show the values estimated from the M^2 curves. The obtained M_s and $1/\chi$ are shown in Fig. 5. The value of P_s is determined as the value of the intersection of natural extrapolation of the M_s curve with the vertical axis in the figure. Our result is almost similar to those reported in Refs. 6 and 7. We also show the M_s^2 and $1/\chi$ vs $T^{4/3}$ plots in the inset of Fig. 5, since the $1/\chi$ and the M^2 of WIFs are predicted to show a non-Fermi-liquid behavior and to obey $T^{4/3}$ relation according to the SCR theory of spin fluctuations. The M_s^2 and $1/\chi$ show the linear relations against $T^{4/3}$ in a relatively wide temperature region, in good agreement with the SCR theory around T_C .

We estimated the Curie constant C and the effective Bohr magneton number P_{eff} by fitting the Curie-Weiss law to the $1/\chi$ from 150 to 300 K. The fitting results are listed in Table II together with the fitting results from 63 to 70 K. In the Table II, P_c is the value determined from $P_{\text{eff}}^2 = 4S(S+1) = P_c(P_c+2)$. From the estimated parameters, we obtained P_{eff}/P_s and T_C/T_0 as 4.8 and 0.085, respectively. We confirmed that these values well satisfy the so-called generalized

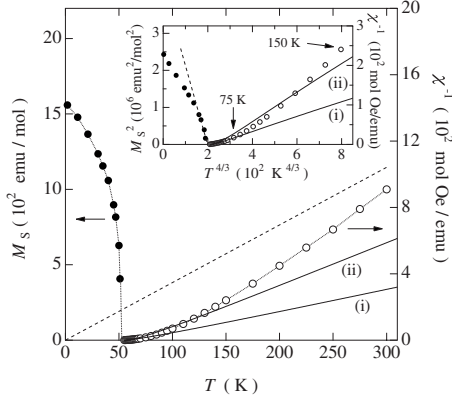


FIG. 5. $M_s(T)$ and $\chi^{-1}(T)$ of LaCoAsO estimated from the Arrott plot. Inset: M_s^2 and χ^{-1} plotted against $T^{4/3}$. Dotted lines are the guides for the eyes. Dashed line is the asymptotic two-dimensional limit of susceptibility. Solid lines represent calculated χ^{-1} 's based on the SCR theory utilizing spin-fluctuation and thermodynamic parameters (see text).

Rhodes-Wohlfarth relation $P_{\text{eff}}/P_s \sim 1.4(T_C/T_0)^{-3/2}$,¹⁹ and can be plotted near the positions of PdCo, CrB₁₂, and Fe_xCo_{1-x}Si (Refs. 13 and 17) as seen in Fig. 6.

Using the spin-fluctuation and the thermodynamic parameters, one can calculate the $1/\chi$ in a paramagnetic state by the SCR theory as

$$y \approx \bar{f}_1 \left(-1 + \frac{1 + \nu y}{c} \int_0^{1/\eta} dz z^3 \left[\ln u - \frac{1}{2u} - \Psi(u) \right] \right), \quad (6)$$

with $y^{-1} = 4\eta^2 T_A \chi / 3$, $u = z(y + z^2)/t$, $\eta = (T_C/T_0)^{1/3}$, $\bar{f}_1 = F_1 P_s^2 / 8 T_A \eta^2$, $\nu = \eta^2 T_A / U$, $c = 0.3353 \dots$, where $\Psi(u)$ is the digamma function, $t = T/T_C$ and U the intra-atomic exchange energy ($\sim 10^4$ K). The results of calculations are shown in Fig. 5 by solid lines. The lines labeled (i) and (ii) are the calculated $1/\chi$ using the values of \bar{F}_1 estimated from Arrott plots and Eq. (5), respectively. The detailed calculation method can be seen in Refs. 12, 13, 20, and 21. The magnetic parameters obtained from the calculated $1/\chi$ (i) and $1/\chi$ (ii) are listed in Table III. Both the lines well reproduce the experimental $1/\chi$ in some region. Especially, the calculated $1/\chi$ (ii) showed good agreement with the experiment in a wide T region rather higher than T_C , indicating that the magnetic property of LaCoAsO can be understood within the Takahashi's developed spin-fluctuation theory just as the cases with MnSi and Fe_xCo_{1-x}Si.^{13,17}

TABLE II. Magnetic parameters of LaCoAsO in a temperature range between 150 and 300 K (upper row) and between 63 and 70 K (bottom row). $P_s = 0.28 \mu_B$.

	θ (K)	P_{eff} (μ_B)	P_c (μ_B)	P_{eff}/P_s	P_c/P_s
High T	98.4	1.34	0.67	4.8	2.4
Low T	58.1	2.99	2.15	10.7	7.7

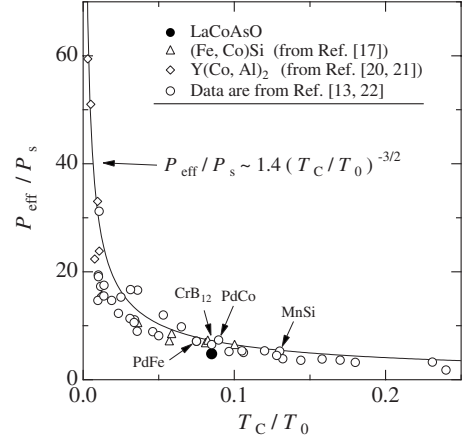


FIG. 6. Generalized Rhodes-Wohlfarth plot. LaCoAsO can be plotted as a closed circle. The other plotted data are reproduced from Refs. 13, 17, and 20–22 and the references therein.

On the other hand, in the narrow T region above T_C , the theoretical $1/\chi$ (i) calculated by using experimentally estimated \bar{F}_1 agrees well with the experiment rather than the $1/\chi$ (ii) as seen in the inset panel, e.g., the value of $P_{\text{eff}}^{\text{call}}$ at 65 K is similar to the experimental one estimated from the fitting between 63 and 70 K. The $1/\chi$ (i) starts to deviate from the experimental $1/\chi$ at about 75 K, which is similar to the temperature where the experimental $1/\chi$ deviates from the $T^{4/3}$ relation. At any rate, our analyses showed that in the vicinity of T_C the SCR theory or Takahashi's developed spin-fluctuation theory quantitatively explains the experimental susceptibility of LaCoAsO. However, above 150 K the experimental susceptibility deviated from the theoretical one as seen in Fig. 5. This result indicates that three-dimensional (3D) spin fluctuations are dominant in a narrow T region near T_C , and the nature of the spin fluctuation should show a cross over from three-dimensional to two-dimensional as increasing T above 150 K. For further discussion on spin fluctuations, we need to cross-check the spin-fluctuation parameters with those estimated from neutron inelastic scattering and NMR measurements, and the latter is now in progress. The extension of the theory for highly anisotropic case is not presently available but under construction.

In the FeAs based high- T_C superconductors, the similar type of spin fluctuations to that in LaCoAsO can be expected

TABLE III. Magnetic parameters from calculated χ (i) and (ii) at 300 K and 65 K by the spin fluctuation theories with $P_s = 0.28 \mu_B$.

	P_{eff} (μ_B)	P_c (μ_B)	P_{eff}/P_s	P_c/P_s
(i)				
300 K	2.43	1.63	8.7	5.8
65 K	2.98	2.14	10.6	7.6
(ii)				
300 K	1.76	1.02	6.3	3.6
65 K	2.31	1.52	8.3	5.4

to be dominant and to be related to the occurrence mechanism of the superconductivity. Recently, Ning *et al.* pointed out that the Co doping into FeAs planes just induces little modulation of local electronic properties of FeAs planes in the case of BaFe_2As_2 ,²³ and the same situation is highly expected in the case of Co doped LaFeAsO .^{7,24} The study of spin fluctuations in LaCoAsO could be expected to inform us of important knowledge to elucidate the nature of superconductivity on the FeAs based high- T_c superconductors.

IV. CONCLUSION

In conclusion, the magnetization of LaCoAsO was found to show a convex curvature as seen in MnSi and $\text{Fe}_{1-x}\text{Co}_x\text{Si}$ when plotted in a form of Arrott plot. By adopting Takahashi's developed spin-fluctuation theory, we estimated spin-fluctuation parameters and calculated $1/\chi$. The good agree-

ment between the experimental and calculated $1/\chi$'s indicates that LaCoAsO is WIF and its characteristics of spin fluctuations can be understood within three-dimensional SCR and Takahashi's spin-fluctuation theories at least around T_C . We indicated a possibility of crossover from 3D to 2D spin fluctuations above 150 K.

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