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Magnetic properties in layered ACo_2Se_2 (A = K, Rb, Cs) with the $ThCr_2Si_2$-type structure

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The magnetic properties of $ThCr_2Si_2$-type single crystals ACo_2Se_2 (A = K, Rb, and Cs) were investigated by means of magnetic susceptibilities and isothermal magnetizations measurements. The ferromagnetic phase transition temperatures are ≈74 and 76 K for A = K and Rb, respectively. A possible antiferromagnetic phase transition occurs around 80 K in susceptibility and a metamagnetismlike behavior is first discovered at a field of ≈3.5 T for $H \parallel ab$ plane in $CsCo_2Se_2$ below 62 K. The susceptibilities in the paramagnetic state obey the modified Curie-Weiss law quite well in all samples. Moreover, the derived effective magnetic moments of the Co atom are about 2.21, 2.04, and 2.04 μ_B/Co for A = K, Rb, and Cs, respectively. The generalized Rhodes-Wohlfarth ratio indicates an itinerant magnetism in this series. Finally, we discussed the magnetic properties in this series within the frameworks of the self-consistent renormalization and Takahashi's theory of spin fluctuations.

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I. INTRODUCTION

The layered compound AT_2X_2-type family shows versatile physical properties including antiferromagnetic (AFM) or ferromagnetic (FM) order,[1] Fe-based superconductivity[2] and heavy fermion behavior.[3] The magnetic properties in this series of compound are very rich. For example, the parent of recently discovered Fe-As based superconductors AFe_2As_2 (A = K, Sr, Ba, and Eu),[4–6] is a collinear antiferromagnetic metal, while (Tl, K, Rb, and Cs)Fe_xSe_2 with Fe vacancies is a block AFM insulator and becomes a superconductor with superconducting temperature ≈30 K with decreasing Fe-vacancy levels.[7] One of a significant common features of Fe-based superconductors is that the parent compounds show an antiferromagnetic phase transition at high temperature which can be suppressed by introducing holes or electrons into conducting layers (FeAs or FeSe). Furthermore, superconductivity also appears in the Ni analogs $BaNi_2As_2$,[8] $SrNi_2As_2$,[9] and KNi_2Se_2,[10] while ferromagnetism or near the ferromagnetism was discovered in Co analogs KCo_2Se_2 (Ref. 11) and $BaCo_2As_2$.[12]

Motivated by the versatility of spin fluctuations involved in AT_2X_2 structure, we turned our attention to the Co-based layered compounds. ACo_2X_2 (A = K, Rb, Cs, and Tl; X = S and Se) has been investigated previously in a polycrystalline sample by Greenblatt's group.[11,13,14] Their results showed that the distance of intralayers or/and interlayers is of vital importance to the electrical and magnetic properties. In this structure the X-T-X (T: transition metals) forms layers, which are built up of edge-sharing TX_4 tetrahedra that extend two dimensionally in the ab plane. For example, KCo_2Se_2 and KCo_2S_2 are ferromagnetic with T_C of around 80 and 120 K, respectively while $CsCo_2Se_2$ is of antiferromagnetic order at low temperatures.[13,15] $TlCo_2Se_2$ is originally thought of as antiferromagnetically ordered with $T_N \approx 90$ K.[11,14] However, the later neutron results showed that it is a noncollinear helical magnet with zero net spontaneous moment, and the Co atoms are ferromagnetically arranged within the ab plane while the magnetic moment of adjacent Co layers are rotated about 121° with respect to each other.[16,17]

The theory of the Stoner-Wohlfarth model,[18] based on the Hartree-Fock mean-field approximation, inadequately explains the temperature dependence of spontaneous magnetization and susceptibility in weakly or nearly ferromagnetic materials due to the near neglect of the spin fluctuation effect. For example, the magnitude of T_C is much smaller than the value predicted in this theory. Moriya and his co-workers developed a self-consistent renormalized (SCR) spin fluctuation theory and succeeded in theoretically reproducing the Curie-Weiss behavior in weakly ferromagnetic metals which was ascribed to the linear growing of $\langle S_L^2\rangle$ with increasing temperature, where $\langle S_L^2\rangle$ is the mean-square of the local amplitude of spin fluctuation.[19] Furthermore, Takahashi expanded the SCR theory by assuming the sum of zero-point and thermal spin fluctuations against temperature is nearly conserved.[20] SCR theory is more applicable to describe the electronic and magnetic properties in weakly or nearly ferromagnetic systems as indicated in an experiment.[21–23]

In this paper, we investigated the temperature dependence of magnetic susceptibility and isothermal magnetization at various temperatures in high quality of single crystals ACo_2Se_2 (A = K, Rb, and Cs), which were grown by the self-flux method. All the samples show metallic luster and metallic conductivity, and the ferromagnetic phase transition temperatures are estimated as ≈74 and 76 K for A = K and Rb, respectively. The susceptibilities obey the modified Curie-Weiss law in the paramagnetic state quite well. The derived effective magnetic moments of the Co atom from susceptibilities are about 2.21 and 2.04 μ_B/Co, and spontaneous moments at the ground state are derived as 0.72 and 0.59 μ_B/Co for samples A = K and Rb, respectively. As a result, the generalized Rhodes-Wohlfarth ratios are estimated as 3.07 and 3.42 for A = K and Rb, indicating itinerant ferromagnetism in this series. The magnetic moments align within the ab plane in samples A = K and Rb. In case of the Cs analog, a metamagnetismlike behavior occurs in a field of ≈3.5 T for the $H \parallel ab$ plane below 62 K, which was not reported in the polycrystalline sample.[13,15] The spin fluctuation effect in this series was discussed within

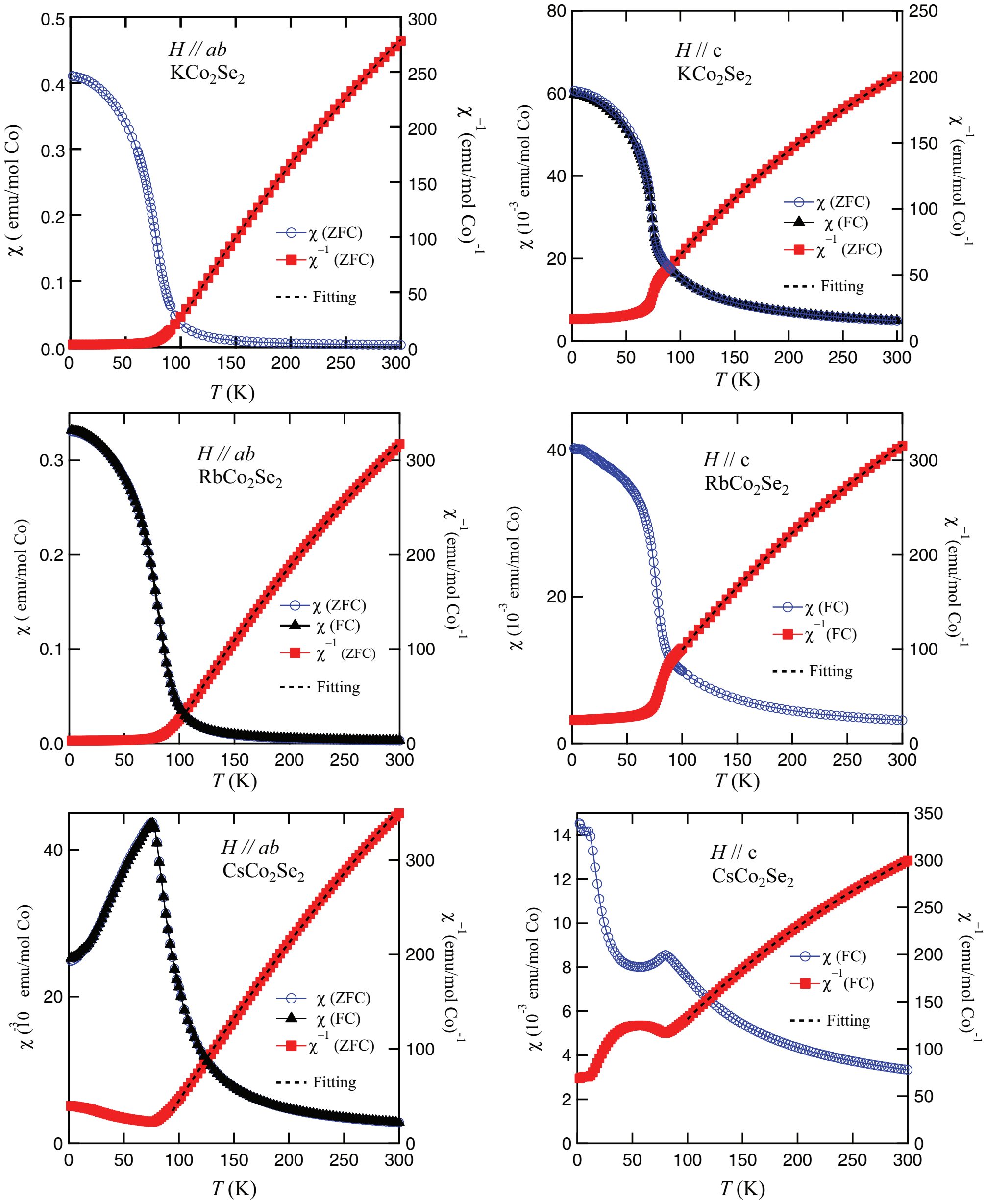

FIG. 1. (Color online) Temperature dependences of the magnetic susceptibilities $\chi(T) = M/H$ and the inverse magnetic susceptibilities ($\chi^{-1} = H/M$) of the ACo_2Se_2 (A = K, Rb, and Cs) in an applied magnetic field of 1 T parallel to the *ab* plane and *c* axis. The dashed lines are the best fitting by the modified Curie-Weiss law in the high temperature region (100 K $\leqslant T \leqslant$ 300 K).

the frameworks of SCR and Takahashi's spin fluctuation theory of weakly ferromagnetic materials.

II. EXPERIMENTS

Single crystals of ACo_2Se_2 (A = K, Rb, and Cs) were grown by the self-flux method. High purity starting materials K_2Se (obtained by the reaction of K metal and Se powder at 573 K for 24 h), K (lump, 99.9%), Co (powder, 99.99%), and Se (powder, 99.999%) were used in the preparation of the single crystals. The stoichiometric amounts of starting materials were mixed carefully and loaded in an Al_2O_3 tube which were sealed inside in an evacuated quartz tube which was coated with carbon to avoid the possible reaction of the quartz tube and the starting materials. We first slowly heated the quartz tube to 673 K and dwelled 5 h. Then the furnace heated slowly up to 973 K (dwelled 24 h) and then heated up to 1323 K and kept at this temperature for 12 h for homogeneity. Finally the furnace slowly cooled to 873 K at a rate of 3 K/h before the furnace was shut down. A typical size of the obtained single crystal is of $\sim 2 \times 3 \times 1$ mm^3. The grown samples have golden luster and are very sensitive to moisture. Therefore, all the above processes are done in the Ar-filled glove box ($H_2O \leqslant 0.1$ ppm and $O_2 \leqslant 0.1$ ppm), and the samples are always kept in the Ar-filled glove box except for a very short time in the air during the preparation of magnetic measurements. The magnetic measurements were performed on the Quantum Design Magnetic SQUID-VSM at Hangzhou Normal University.

TABLE I. Magnetic parameters of ACo_2Se_2 (A = K, Rb, and Cs). χ_0, C, θ, P_s, and P_{eff} are temperature-independent parts of χ (emu/mol Co), Curie constant C (10^{-3} emu/mol Co K), Curie temperature θ (K), spontaneous magnetization at ground state (μ_B/Co atom) and effective moment (μ_B/Co atom), respectively. We also calculated the ratio of P_{eff}/P_s and the value of P_c, where $P_c(P_c + 2) = P_{\mathrm{eff}}^2$.

Sample	χ_0	C	θ	P_{eff}	P_c	P_s	P_{eff}/P_s
K ($H \parallel ab$ plane)	0.8	0.61	85.2	2.21	1.41	0.72	3.07
K ($H \parallel c$ axis)	2	0.84	38.2	2.59	1.78		
Rb ($H \parallel ab$ plane)	0.7	0.52	88.5	2.04	1.27	0.59	3.46
Rb ($H \parallel c$ axis)	1	0.57	37.4	2.14	1.36		
Cs ($H \parallel ab$ plane)	0.5	0.52	75.8	2.04	1.27	0.52	3.92
Cs ($H \parallel c$ axis)	1	0.58	7.5	2.15	1.37		

III. RESULTS AND DISSCUSSION

A. Susceptibility

Figure 1 shows the temperature dependence of magnetic susceptibility ($\chi = M/H$) and inverse magnetic susceptibility ($\chi^{-1} = H/M$) for single crystals ACo_2Se_2 (A = K, Rb, and Cs) in an applied magnetic field of 1 T parallel to the ab plane or c axis. First, we discuss the χ(T) for A = K and Rb single crystals. The quick increase of susceptibilities below at about 80 K when further decreasing the temperature is due to the occurrence of a long range of ferromagnetic order, and the susceptibilities tend to a saturated value of about 0.4 emu/mol Co at low temperatures. This saturated value is only 15% of that in the polycrystal as previous reported,[11] indicating the high quality of our single crystals. At high temperatures, χ can be well fitted by the modified Curie-Weiss law:

$$\chi = \chi_0 + \frac{C}{T - \theta}, \tag{1}$$

where χ_0 denotes the temperature-independent term, C the Curie constant, and θ the paramagnetic Curie temperature. The above parameters, derived in the temperature from 100 to 300 K for $H \parallel ab$ plane or $H \parallel c$ axis, are listed in Table I. The Curie constants are nearly independent of samples with the value of around 0.6 emu K/mol, and the effective magnetic moment P_{eff} about 2.4 μ_B. This value is smaller than the theoretical value of 3.87 μ_B for a free Co^{2+} ion, suggesting the Co ion is of an itinerant character. The Curie temperature θ_{ab}, derived for $H \parallel ab$ plane, is almost unchanged in the samples A = K, Rb, and Cs. However, the value of θ_c, derived for $H \parallel c$ axis, is nearly half of θ_{ab} for samples A = K and Rb, and in the $CsCo_2Se_2$ case, θ_{ab}/θ_c is increased to 10. The positive value of θ_c and θ_{ab} means a ferromagnetic interaction between the adjacent interlayer Co_2Se_2 layers or within the same layer is dominant. A large difference of θ derived from $H \parallel ab$ plane and c axis indicates that the strength of interaction of Co atoms within the layer is stronger than it is between the layers. On the other hand, the susceptibility in $CsCo_2Se_2$ displays a cusplike peak around 80 K and seems due to an antiferromagnetic phase transition, which contrasts to the saturated behavior discovered in samples A = K and Rb. This series of samples A = K, Rb, and Cs belong to a soft ferromagnet because there is no apparent hysteresis in the M (H) curves as shown in Fig. 2.

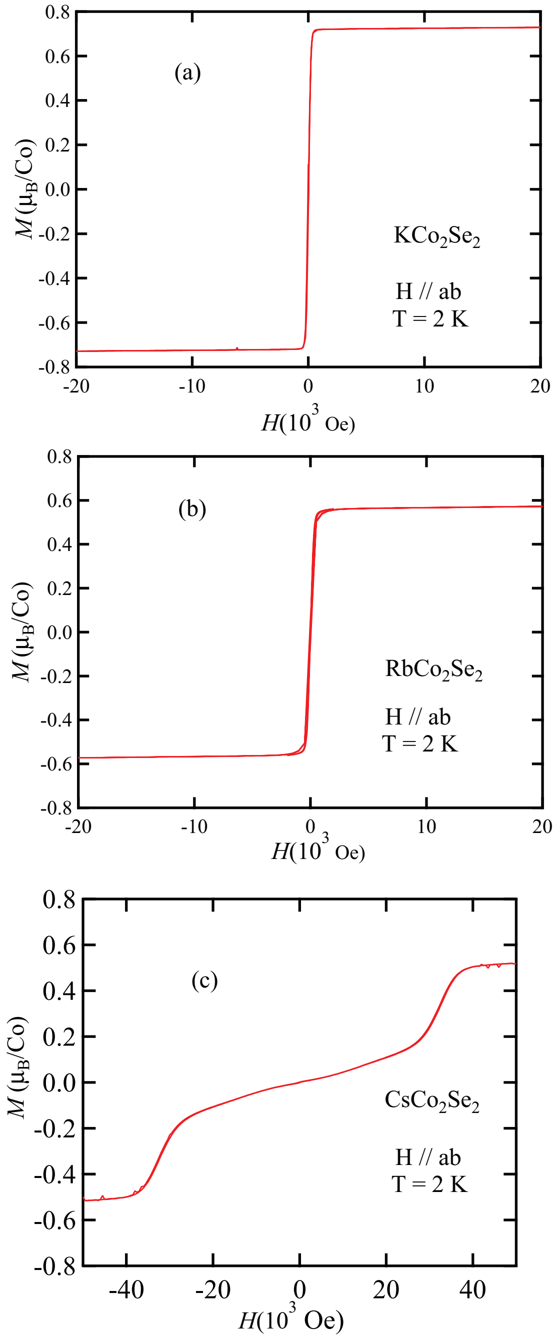

FIG. 2. (Color online) Hysteresis loops for ACo_2Se_2 (A = K, Rb, and Cs) measured in an applied magnetic field $H \parallel ab$ plane at 2 K.

B. Isothermal magnetization

Figure 3 shows the results of magnetic measurements for single crystals of KCo_2Se_2 and $RbCo_2Se_2$ in forms of

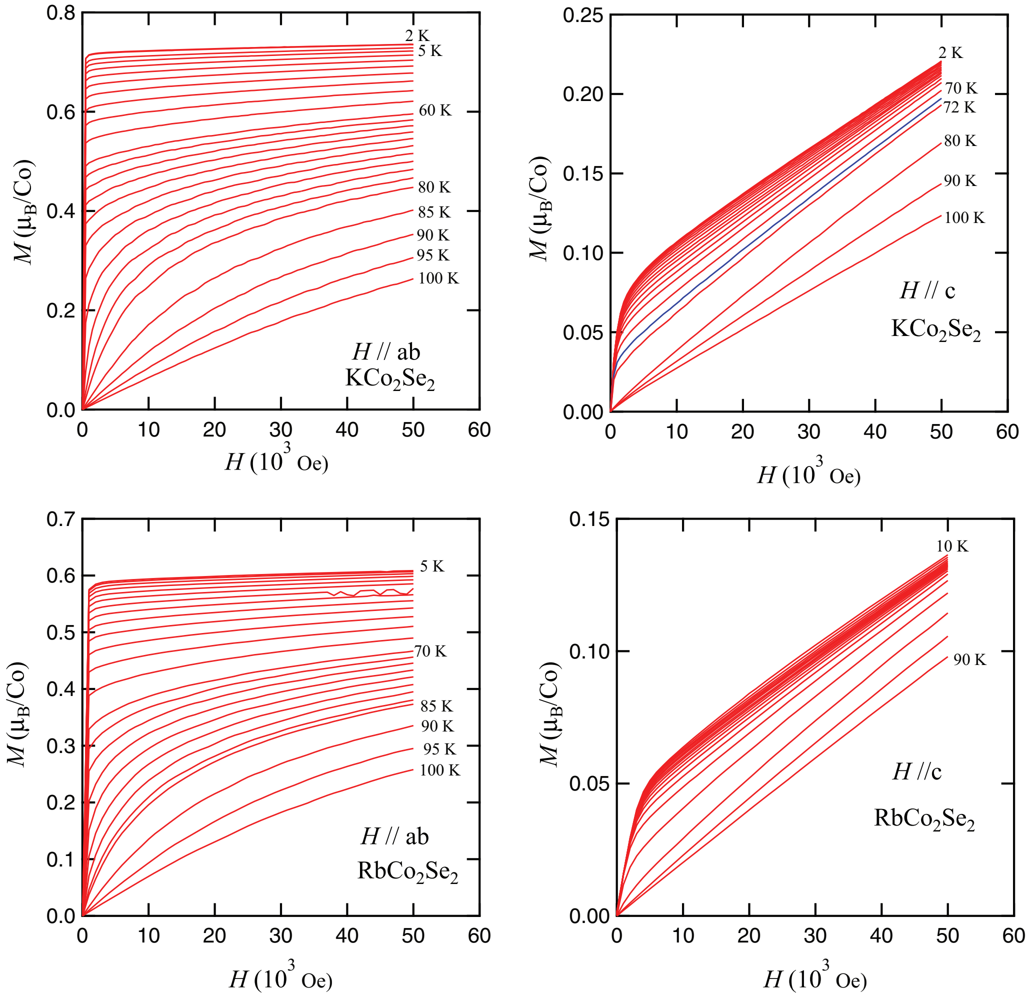

FIG. 3. (Color online) Isothermal magnetization measurements for $A = \text{K}$ and Rb single crystals at temperatures from 2 to 300 K in an applied magnetic field $H \parallel ab$ plane or $H \parallel c$ axis. Not all data are shown. The steps of the temperature are 2 or 5 K in the figure.

isothermal magnetization $M(H)$ curves and the corresponding Arrott plots at different temperatures in an applied magnetic field $H \parallel ab$ plane or $H \parallel c$ axis. The magnetization goes quickly to saturated magnetic moments $0.75\mu_B$ and $0.60\mu_B$ for KCo_2Se_2 and RbCo_2Se_2 at $H \parallel ab$ plane, respectively. The magnetic moment increases with increasing magnetic field; however, there is no tendency to saturation for $H \parallel c$ axis up to the field of 5 T. Figure 4 displays the magnetic field dependence of the magnetic moment at various temperatures for single crystal CsCo_2Se_2. A metamagnetismlike behavior occurs at the field of 3 to 4 T at a field $H \parallel ab$ plane and below the temperature of around 62 K in the $M(H)$ curve. The saturation moment is $\approx 0.55\ \mu_B/\text{Co}$ at the field of 5 T. No metamagnetismlike behavior was discovered for $H \parallel c$ axis, but a slight kink was discovered in a similar field region at 10 K as shown in the inset of Fig. 4. In the polycrystal $\text{TlCo}_2\text{Se}_{2-x}\text{S}_x$ ($x = 1.3$ and 1.5), similar behavior was discovered in the field of ≈ 1 T which was ascribed to the possible collapse of the helical structure at high magnetic field.[24]

According to Landau theory, the free energy F can be expressed as the expansion of an order parameter. Therefore, from $H = \partial F/\partial M$, we obtain $H = aM(T,H) + bM^3(T,H)$ or as often written as $M^2(T,H) = M^2(T,0) + BH/M(T,H)$ by neglecting the sixth and higher terms of the expansion, resulting in the so-called Arrott plot. Figure 5 shows the Arrott plots for single crystals $A = \text{K}$ and Rb for $H \parallel ab$ plane or $H \parallel c$ axis. As shown in Fig. 5, a good linear behavior in the Arrott plot of KCo_2Se_2 and RbCo_2Se_2 only exists at low temperatures and at high magnetic field side for $H \parallel ab$ plane. In the Arrott plots, we can estimate the value of the spontaneous magnetic moment by extrapolating the linear relation to a positive intersection on the y axis. It is clearly shown in Figs. 5(a) and 5(c) that the spontaneous magnetic moments of $A = \text{K}$ and Rb single crystals are aligned within the ab plane since the linear extrapolation to the y axis for $H \parallel c$ axis is negative at the lowest temperature of 2 K which means no spontaneous magnetic moment along this direction.

Generally, T_C can be obtained by using Arrott plots since the linear extrapolation passes through the origin at T_C. However, the curves show considerable deviation from linearity at high temperatures in our case which makes it difficult to estimate the T_C in this system. In Takahashi's theory, the zero-point and thermal spin fluctuation amplitude for a weak ferromagnet is nearly conserved and the fourth order expansion coefficient vanishes at the critical

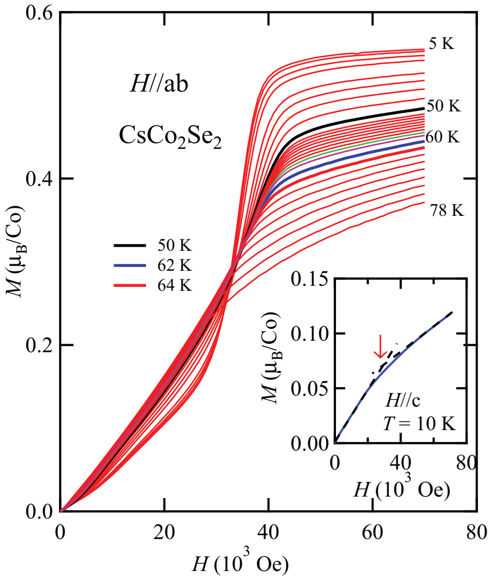

FIG. 4. (Color online) Isothermal magnetization measurements for the $A = \mathrm{Cs}$ sample at various temperatures for $H \parallel ab$ plane. Inset: Isothermal magnetization measurements for $H \parallel c$ axis at 10 K. The steps of the selected temperature are 1, 2, or 5 K in the figure and not all data are shown.

temperature.[25] Therefore, the magnetization near T_C obeys the relation:

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

where $h = 2\ \mu_B H$, $P = 2M(T)/N_0$. N_0 is the number of magnetic atoms in the unit cell, μ_B the Bohr magneton, and T_A the dispersion of the static magnetic susceptibility in wave-vector space. This result gives a more simple relation $M^4 \propto H/M$ near T_C. Figure 6 shows M^4 dependence of H/M, indicating a good linear relation behavior in a wider range of temperatures for crystals of $A = \mathrm{K}$ and Rb. Such behavior is also discovered in the case of CsCo$_2$Se$_2$ at high magnetic fields shown in Fig. 7. Therefore, we estimated T_C in this series as 74 (K), 76 (Rb), and 62 K (Cs) by linear extrapolation at the temperature which passes through the origins. In Takahashi's theory, the overall linearity of the Arrott plot is due to a small value of $\eta = (T_C/T_0)^{1/3}$, where T_0 is energy width of the dynamical spin fluctuation spectrum. As a result, very good linearities are observed experimentally in Arrott plot curves in Ni$_3$Al ($\eta = 0.25$)[26] and Sc$_3$In ($\eta = 0.22$),[27] while a relatively larger value of η gives worse linearities in Arrott plot curves as observed in MnSi ($\eta = 0.58$),[28] LaCoAsO($\eta \approx 0.46$),[29] and Fe$_3$GeTe$_2$ ($\eta \approx 0.46$).[30] The larger η indicates the existence of sixth-order nonlinear mode-mode coupling

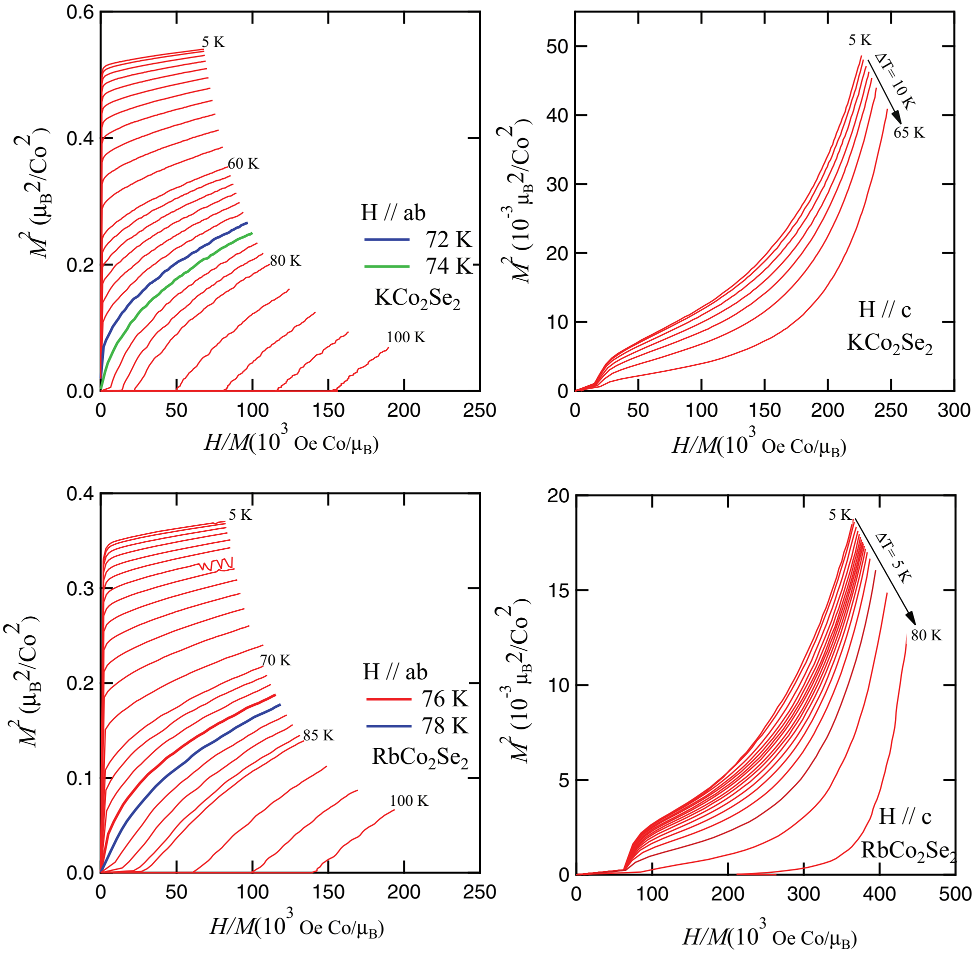

FIG. 5. (Color online) M^2 vs H/M (Arrott plots) for $A = \mathrm{K}$ and Rb samples at various temperatures in an applied magnetic field $H \parallel ab$ plane and $H \parallel c$ axis, respectively. The steps of the selected temperatures are 2 or 5 K in the figure.

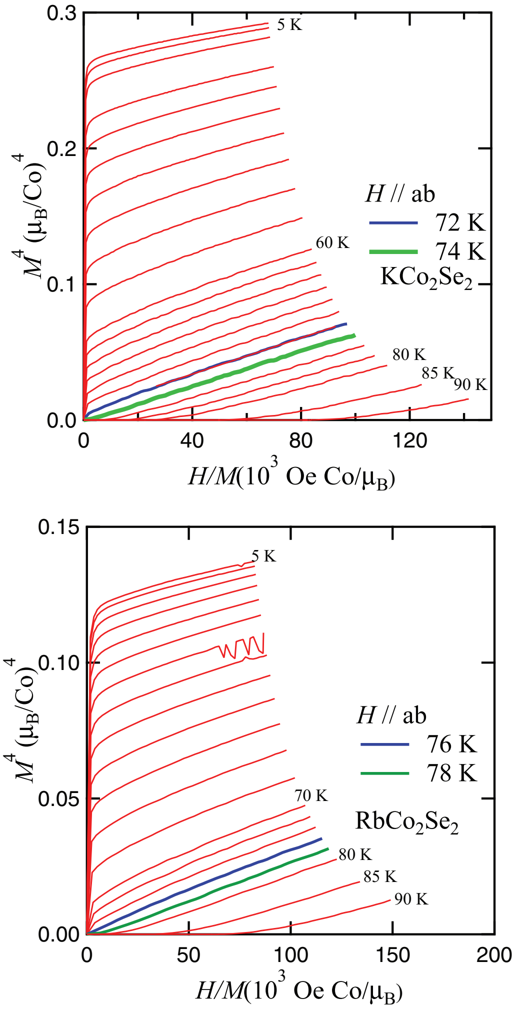

FIG. 6. (Color online) M^4 vs H/M for $A = $ K and Rb single crystals at various temperatures for $H \parallel ab$ plane. We measured the isothermal magnetization from 2 to 300 K and only a part of the data are shown here for clarity. The steps of the temperatures are 2 or 5 K in the figure.

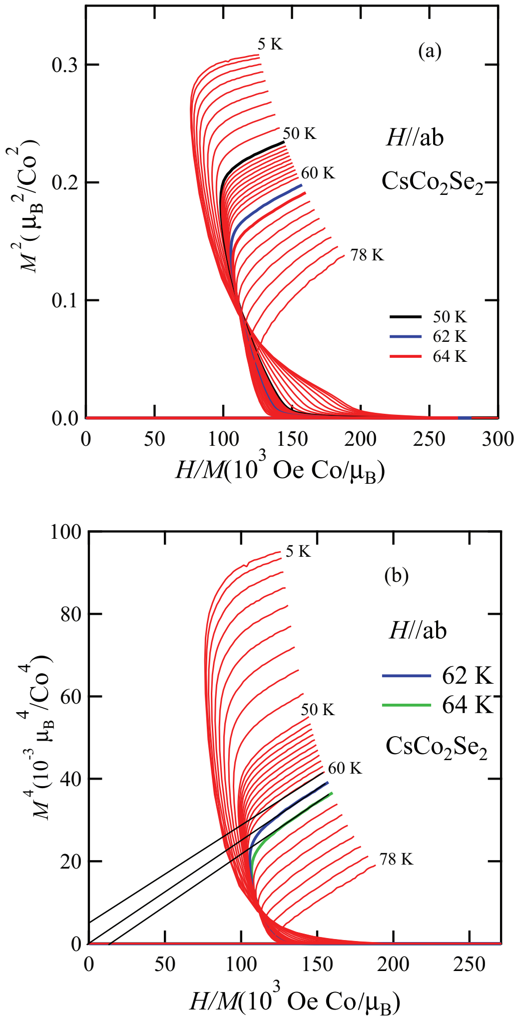

FIG. 7. (Color online) (a) M^2 vs H/M and (b) M^4 vs H/M at various temperatures for the $A = $ Cs sample for $H \parallel ab$ plane. The steps of the selected temperatures are 1, 2, or 5 K in the figure. The three solid lines are guides to eyes to indicate that the metamagnetism survives below 62 K in CsCo$_2$Se$_2$.

among spin fluctuation modes in our case.[19] We estimated η for our single crystals $A = $ K, Rb, and Cs are 0.64, 0.64, and 0.57, respectively by using T_C and T_0 listed in Table II below. On the other hand, the spontaneous magnetic moment $M_0(T)$ and the reciprocal initial magnetic susceptibility $1/\chi_{\text{initial}}$ $(=\lim_{H\to 0} H/M)$ are derived from the linear extrapolation in curves of M^4 vs H/M. From the estimated parameters above, we obtained the values of P_{eff}/P_s and T_C/T_0. These values satisfy the so-called generalized Rhodes-Wohlfarth relation $P_{\text{eff}}/P_s = 1.4(T_C/T_0)^{-2/3}$ and Deguchi-Takahashi plot quite well as shown in Fig. 8.[25,31]

The spin fluctuation spectrum, characterized as T_A and T_0, plays an important role in Moyria and Takahashi's theory for the weakly ferromagnet.[19,25] These two important parameters dominate physical properties at the ground state and finite temperatures. Furthermore, another important spin-fluctuation parameter $\bar{F}_{10}$ also can be derived from the Arrott plot which

TABLE II. Spin-fluctuation parameters of ACo$_2$Se$_2$ ($A = $ K, Rb, and Cs). T_C and $\bar{F}_{10}$ are derived from Arrott plots. T_0 and T_A are calculated according to SCR theory. The T_0^* and T_A^* are the best fits to the initial magnetic susceptibility data on Takahashi's theory. The estimated errors are shown in the brackets.

Sample (A)	T_C (K)	T_0 (10^2 K)	T_A (10^3 K)	$\bar{F}_{10}$ (10^4 K)	$\bar{F}_{10}^*$ (10^4 K)	T_0^* (10^2 K)	T_A^* (10^3 K)
K	74	2.82	1.83	3.15(0.01)	2.75(0.1)	3.06(0.1)	1.78(0.1)
Rb	76	2.93	2.22	4.49(0.02)	4.10(0.1)	4.23(0.1)	2.55(0.1)
Cs	62	3.48	2.58	5.10(0.02)	4.00(0.2)	4.02(0.1)	2.46(0.1)

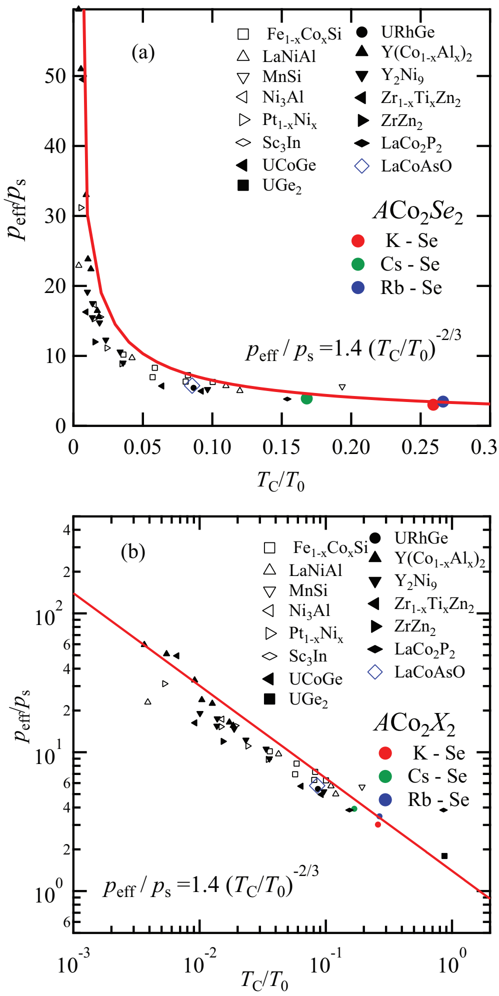

FIG. 8. (Color online) (a) Generalized Rhodes-Wohlfarth plot. $A\mathrm{Co}_2\mathrm{Se}_2$ are plotted as closed circles. (b) Deguchi-Takahashi plot. The other data are reproduced from Refs. [19,21,27–29,33–40] and references therein Ref. [25].

is the coefficient of the M^4 term of the Landau expansion of free energy and is usually expressed in kelvin units as

$$\bar{F}_{10} = N_A^3(2\mu_B)^4/\xi K_B, \qquad (3)$$

where N_A and k_B are Avogadro's number and the Boltzmann constant, respectively, and ξ is the slope of the Arrott plot at T_C. Therefore, $\bar{F}_{10}$ can be determined uniquely from the slope of the Arrott-plot at T_C. Although the curves of the Arrott plot (M^2 vs H/M) don't have a perfect linearity, it is reasonable to estimate the gradient at the temperatures of T_C in a narrower high magnetic field region. We estimated $\bar{F}_{10}$ from a high field region of the M^2 vs H/M curve at T_C. In Takahashi's theory, $\bar{F}_{10} = \frac{4T_A^2}{15T_0}$, and hence gives the relations below:[20]

$$\left(\frac{T_C}{T_0}\right)^{5/6} = \frac{\sqrt{30C_z}\,M_0(0)^2}{40C_{4/3}}\left(\frac{\bar{F}_{10}}{T_C}\right)^{1/2}, \qquad (4)$$

$$\left(\frac{T_C}{T_A}\right)^{5/3} = \frac{M_0(0)^2}{20C_{4/3}}\left(\frac{2T_C}{15C_z\bar{F}_{10}}\right)^{1/3}, \qquad (5)$$

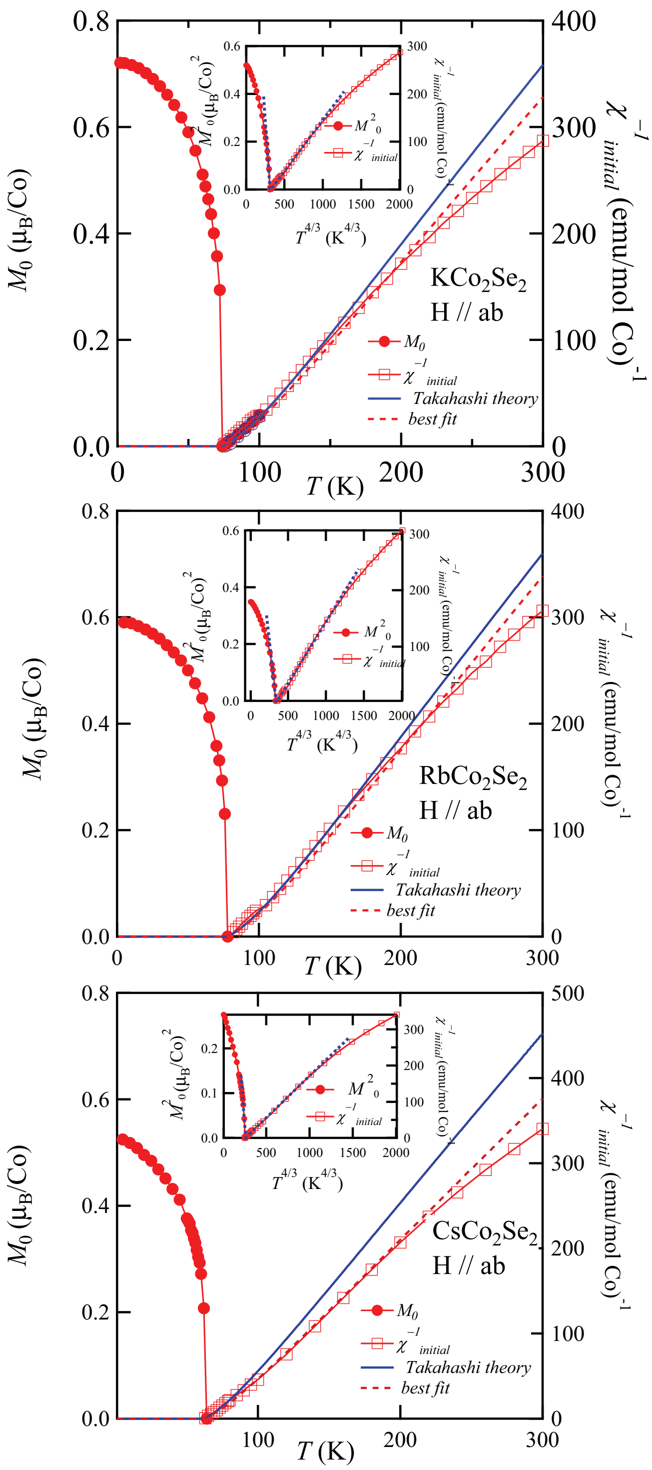

FIG. 9. (Color online) Temperature dependence of spontaneous magnetization (M_0 vs T) in $A\mathrm{Co}_2\mathrm{Se}_2$ (A = K, Rb, and Cs) and the initial susceptibilities for $H \parallel ab$ plane. Insets: M_0^2 and $\chi^{-1}_{\rm initial}$ against $T^{4/3}$.

where the integral constants $C_{4/3}$ and C_z are parameters to calculate the thermal amplitude of the spin fluctuation and the values are 1.006 and 0.50, respectively, for the weakly ferromagnetic limit, and $M_0(0)$ the spontaneous magnetic moment at the ground state. The detailed spin fluctuation parameters are listed in Table II.

Using the spin fluctuation and thermal dynamic parameters, we calculated the $1/\chi_{\rm initial}$ in a paramagnetic state by the SCR

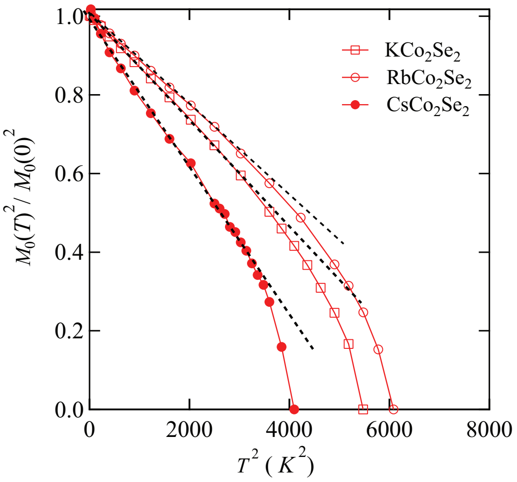

FIG. 10. (Color online) Temperature dependence of spontaneous magnetization in single crystals ACo_2Se_2 (A = K, Rb, and Cs). The dashed lines are the best fits to the data at low temperatures.

theory[19] as

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

with $y^{-1} = 4\eta^2 T_A \chi/3$, $u = z(y + z^2/t)$, $\eta = (T_C/T_0)^{1/3}$, $\bar{f}_1 = \bar{F}_{10} P_s^2 / 8 T_A \eta^2$, $v = \eta^2 T_A / U$, $c = 0.3353$, where $\Psi(u)$ is the digamma function, $t = T/T_C$, and U the intra-atomic exchange energy. Therefore the calculated temperature dependence of the inverse magnetic susceptibilities for samples of A = K, Rb, and Cs by using the parameters T_0, T_A, $\bar{F}_{10}$, and M_0 obtained from the experiment on Eq. (6) are shown in solid lines in Fig. 9. The solid lines show clear deviations at high temperatures but agree well with the experimental data at low temperatures. If we allow the parameters T_0, T_A, and $\bar{F}_{10}$ to be changeable we can obtain the best fits to each sample as shown by dashed lines in the insets of Fig. 9. As a result, the parameters of T_0^*, T_A^*, and $\bar{F}_{10}^*$ are a little larger than the values obtained from the Arrott plots, however, in the same order of magnitude. In the SCR theory and Takahashi's theory, the squared spontaneous moment M_0^2 and the initial susceptibility $\chi_{\text{initial}}^{-1}$ have a relation of $T^{4/3}$ near the T_C as shown in the insets of Fig. 9. Very near T_C, the coefficients of $T^{4/3}$ are given as $a_c = 1.27$ for a weakly ferromagnetic limit.[20] We obtained the coefficients of $T^{4/3}$ for A = K, Rb, and Cs, which are 1.27, 1.31, and 1.03, respectively by fitting the data near T_C. The results satisfied the theory very well except for the Cs case which is possibly due to its very narrow $T^{4/3}$ temperature region. This means ACo_2Se_2 (A = K, Rb) belong to weakly ferromagnetic materials. In Takahashi's theory, T^2 dependence of the squared spontaneous moment at low temperatures is described in terms of T_A by neglecting the spin-wave contribution at low temperatures and obeys the formula:[32]

$$\left[\frac{M_0(0,T)}{M_0(0)}\right]^2 = 1 - \frac{50.4}{M_0(0)^4}\left(\frac{T}{T_A}\right)^2. \tag{7}$$

A good linearity is shown in curves of $[\frac{M_0(T)}{M_0(0)}]^2$ against T^2 at low temperatures as displayed in Fig. 10. As results, the fitting by using formula (7) gives the values of T_A, which are 1193, 1900, and 1828 K for samples of A = K, Rb, and Cs, respectively. These values are very close to those derived from $\bar{F}_{10}$.

The parameters of T_0 and T_A derived from the Arrott plots or the initial magnetic susceptibility calculated on the SCR theory and Takahashi's theory are comparable with each other. Similar results were reported in the LaCoAsO system.[29] It strongly indicates that the ferromagnetic spin fluctuations in Co analogs related to the newly discovered Fe-based superconductors can be understood within the frameworks of the SCR theory and Takahashi's theory. The study of spin fluctuations in ACo_2Se_2 (A = K, Rb, and Cs) could give more information to shed light on the nature of superconductivity on Fe-based superconductors.

IV. CONCLUSION

In summary, we investigated the temperature dependence of the magnetic susceptibility and isothermal magnetization for the single crystals ACo_2Se_2 (A = K, Rb, and Cs), which were grown by the self-flux method. A ferromagnetic transition occurs at ≈74 and 76 K for A = K and Rb, respectively. $CsCo_2Se_2$ is possibly antiferromagnetically ordered at 80 K and a metamagnetismlike behavior occurs at about 3.5 T for $H \parallel ab$ axis in $CsCo_2Se_2$ below 62 K. The magnetic susceptibility obeys the modified Curie-Weiss law in the paramagnetic state quite well. The Arrott plots show linear behavior at low temperatures and nonlinear behavior in high temperatures. A good linear relation is realized in the curves of M^4 vs H/M in a quite broad temperature region, which is supported by a large η in Takahashi's theory. The estimated spontaneous magnetic moments at zero temperature are 0.72, 0.58, and 0.52 μ_B for A = K, Rb, and Cs (in a high magnetic state), respectively. The accordingly generalized Rhodes-Wohlfarth ratios are 3.07, 3.46, and 3.92, indicating a moderate itinerant ferromagnetism in this system. The spin fluctuation in this system can be understood in the frameworks of the SCR theory and Takahashi's theory of spin fluctuations.

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