AIP | Journal of Applied Physics

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Citation: J. Appl. Phys. **109**, 113911 (2011); doi: 10.1063/1.3594752

View online: http://dx.doi.org/10.1063/1.3594752

View Table of Contents: http://jap.aip.org/resource/1/JAPIAU/v109/i11

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JOURNAL OF APPLIED PHYSICS **109**, 113911 (2011)

3D-Heisenberg ferromagnetic characteristics in $CuCr_2Se_4$

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(Received 16 January 2011; accepted 26 April 2011; published online 6 June 2011)

The critical properties of the spinel selenide $CuCr_2Se_4$ around T_C have been investigated by the modified Arrott plot, Kouvel–Fisher method, and critical isotherm analysis. Reliable critical exponents $\beta = 0.372 \pm 0.007$, $\gamma = 1.277 \pm 0.044$, and $\delta = 4.749 \pm 0.016$ with $T_C = 430$ K are obtained. Based on these critical exponents, the magnetization-field-temperature (*M-H-T*) data around T_C collapses into two independent curves obeying the single scaling equation $M(H, \varepsilon) = \varepsilon^{\beta} f_{\pm}(H/\varepsilon^{\beta+\gamma})$. Moreover, the critical exponents are confirmed by the field dependence of the magnetic entropy change relation $\Delta S_M|_{T=T_C} \propto H^n$ with $n = 1 + (\beta - 1)/(\beta + \gamma)$. The obtained critical exponents are in good agreement with the prediction of the 3D-Heisenberg model except that γ is smaller than the theoretical value. The smallness of γ may be due to the delocalization of holes produced by Se^{1-}, which gives evidence for the model of the ferrimagnetic hybridization between localized electrons of the Cr^{3+} ions and delocalized holes of the Se 4*p* band proposed for $CuCr_2Se_4$. © *2011 American Institute of Physics*. [doi:10.1063/1.3594752]

I. INTRODUCTION

The chromium-based selenide $CuCr_2Se_4$ is well known for the highest ferromagnetic-paramagnetic (FM-PM) phase transition temperature $T_C = 430$ K among chromium spinel chalcogenides.[1,2] This compound is also paid considerable attention because of the theoretical prediction of highly spin-polarization and exhibition of significant magneto-optic Kerr rotation, which makes it a candidate material for new magnetic devices.[3,4] $CuCr_2Se_4$ is an itinerant ferrimagnetic metal with measured moments of $\sim 5\mu_B$ in a unit cell.[5,6] Recent band structure calculations demonstrate that $CuCr_2Se_4$ is almost half-metallic,[4,6–8] where the density of states for spin-down electrons can be fully suppressed with cadmium doping to realize a perfect half-metallic situation.[8]

The origin of the magnetism for $CuCr_2Se_4$ has been in controversy for a long time. Lotgering and Stapele invoked a double exchange (DE) model, which assumed a electronic configuration of $Cu^{1+}(Cr^{3+}Cr^{4+})Se^{2-}$.[9] In this model, the ferromagnetism and metallic conductivity were established through DE between Cr^{3+} and Cr^{4+}, which gave a perfect explanation to magnetic and transport behaviors. However, neutron diffraction studies indicated that each chromium was in the Cr^{3+} state.[5] Later, the itinerant ferromagnetism was explained by Goodenough,[10] who proposed a divalent Cu^{2+} whose moments were antiparallel to the ferromagnetic moments of Cr^{3+} via 90° superexchange through the completely filled Se 4*p* states. The x-ray magnetic circular dichroism (XMCD) measurements confirmed the Cr^{3+} state.[11] However, it was found almost monovalent copper and a delocalized hole in the Se 4*p* band with a magnetic moment antiparallel to moments of Cr^{3+} ions. Then, Saha-Dasgupta *et al*. suggested the ferrimagnetic hybridization between localized electrons of Cr^{3+} ions and delocalized holes of Se 4*p* band resulting in a hole-mediated exchange, i.e., $Cu^{1+}Cr^{3+}_{(\uparrow)}Se^{1-}_{(\downarrow)}Cr^{3+}_{(\uparrow)}Se^{2-}_3$.[4,6,8] This model has been confirmed by density functional calculations and magneto-optical study, which indicated the appearance of a hybridization-induced humplike structure at the Fermi energy only for the spin-up states.[12]

As is well known, a study of critical behaviors is beneficial to understand magnetic properties. In this work, critical behaviors of $CuCr_2Se_4$ were analyzed. Four kinds of different theoretical models, mean-field ($\beta = 0.5$), 3D-Heisenberg ($\beta = 0.365$), 3D-Ising ($\beta = 0.325$), and tricritical mean field ($\beta = 0.25$) in view of the varied critical exponent β from 0.1 to 0.5 were tried to explain the critical properties.[13,14] Reliable critical exponents β, γ, and δ were obtained, and confirmed by the scaling hypothesis and magnetic entropy change relation. It is found that the $CuCr_2Se_4$ is in best accordance with the 3D-Heisenberg model, except that γ is smaller than the theoretical value. The ferrimagnetic hybridization between localized electrons of Cr^{3+} ions and delocalized holes of Se 4*p* band should be responsible for the smallness of γ, which supports the ferrimagnetic hybridization model.

II. EXPERIMENT

A polycrystalline sample of $CuCr_2Se_4$ was prepared by the solid-state reaction method.[15] The powder x-ray diffraction proves that the sample is single phase with cubic cell belonging to the space group $Fd\bar{3}m$. The magnetic properties were measured with a commercial superconducting quantum interference device magnetometer (Quantum Design MPMS). The sample was processed to ellipsoid shape and the field was applied along the long axis to decrease the

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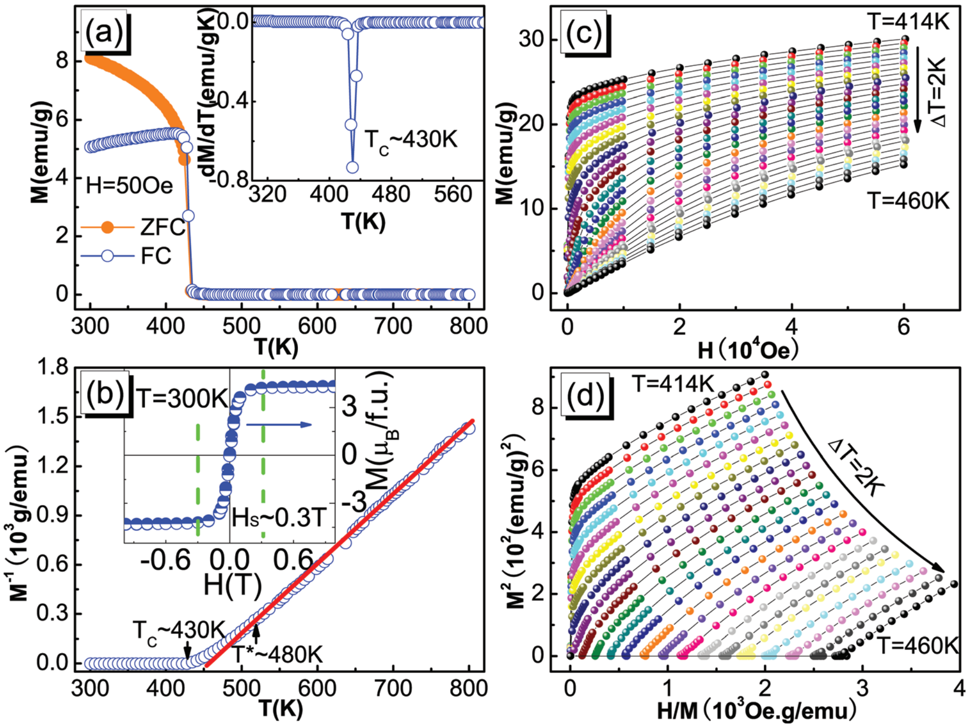

FIG. 1. (Color online) (a) The temperature dependence of magnetization under ZFC and FC for $CuCr_2Se_4$ (the inset plots the dM/dT vs T); (b) the inverse magnetization $M^{-1}(T)$ [the inset is the $M(H)$ at 300 K]; (c) the isothermal magnetization around T_C; (d) the Arrott plot: isotherms of M^2 vs H/M.

demagnetizing field. The isothermal magnetization was performed after the sample was heated well above the Curie temperature T_C for more than ten minutes to ensure every curve was an initial magnetizing curve.

III. RESULTS AND DISCUSSION

Figure 1(a) shows the temperature dependence of magnetization under zero-field-cooling (ZFC) and field-cooling (FC) for $CuCr_2Se_4$. In order to reveal intrinsic magnetic properties of this system, the magnetization was measured under a low field of 50 Oe. A PM-FM phase transition occurs at T_C. The λ shape of the ZFC and FC curves implies glass-clusters in this system. The inset of Fig. 1(a) plots the derivation of magnetization for the ZFC curve as a function of the temperature. The dM/dT versus T at low magnetic field enables us to determine precise T_C because of the avoidance of a great influence of domain structures in the FM phase.[16] The minimum of dM/dT corresponds to T_C. As can be seen, T_C approximates 430 K for $CuCr_2Se_4$, consistent with previous report.[1] Meanwhile, T_C can be also determined by the inverse magnetization. Figure 1(b) plots the $M^{-1}(T)$ for $CuCr_2Se_4$. An inflection is found at 430 K, corresponding to T_C. However, M^{-1} deviates from linear behavior around 480 K, which implies the presence of critical fluctuations above T_C.[17] In fact, T_C is usually affected by the external magnetic field in such kind of ferromagnet, such as $CuCr_2Te_4$ and $CdCr_2Se_4$.[16,18] Thus, the exact T_C, as well as other critical parameters, should be determined by the critical analysis.

The isothermal magnetization at temperatures around the critical point ($|(T_C - T)/T_C| < 0.1$) is given in Fig. 1(c) for the analysis of critical behaviors. The inset of Fig. 1(b) plots the $M(H)$ at 300 K in the FM phase, indicating that $CuCr_2Se_4$ is a soft ferromagnet with saturation field $H_S \approx 0.3$ T. As we know, the critical parameters can be determined easily by the analysis of the Arrott plot at temperatures around the critical point.[19,20] According to the Landau theory of phase transition, the Gibbs free energy G can be expressed in terms of the order parameter M as

$$G(T,M) = G_0 + aM^2 + bM^4 - MH. \quad (1)$$

The coefficients a and b are temperature-dependent.[21] For the condition of equilibrium ($\partial G/\partial M = 0$, i.e., energy minimization), the magnetic equation of state evolves into

$$H/M = 2a + 4bM^2. \quad (2)$$

Generally, M^2 versus H/M should be a series of parallel straight lines in the high field range in the Arrott plot. The intercept of the M^2 as a function of H/M on the H/M axis is negative/positive below/above T_C. The line of M^2 versus H/M at T_C should pass through the origin. According to the criterion proposed by Banerjee,[22] the order of the magnetic transition can be determined from the slope of the straight line: the positive slope corresponding to the second order transition while the negative slope to the first order one. Figure 1(d) depicts the Arrott plot around T_C for $CuCr_2Se_4$. Apparently, the positive slop of the M^2 versus H/M implies that the PM-FM transition is a second order one. However, all curves in the Arrott plot are nonlinear and show curvature downward even in the high field region, which indicates that $\beta = 0.5$ and $\gamma = 1.0$ are not satisfied according to the Arrott–Noakes equation of state $(H/M)^{1/\gamma} = (T - T_C)/T_C + (M/M_1)^{1/\beta}$.[23] In other words, the Landau mean-field theory with $\beta = 0.5$ and $\gamma = 1.0$ is not valid for $CuCr_2Se_4$. Hence, the modified Arrott plot should be employed to obtain correct critical parameters.

As for a second order magnetic phase transition, its critical behaviors can be studied through a series of critical exponents. In the vicinity of a second order magnetic phase transition, the divergence of correlation length

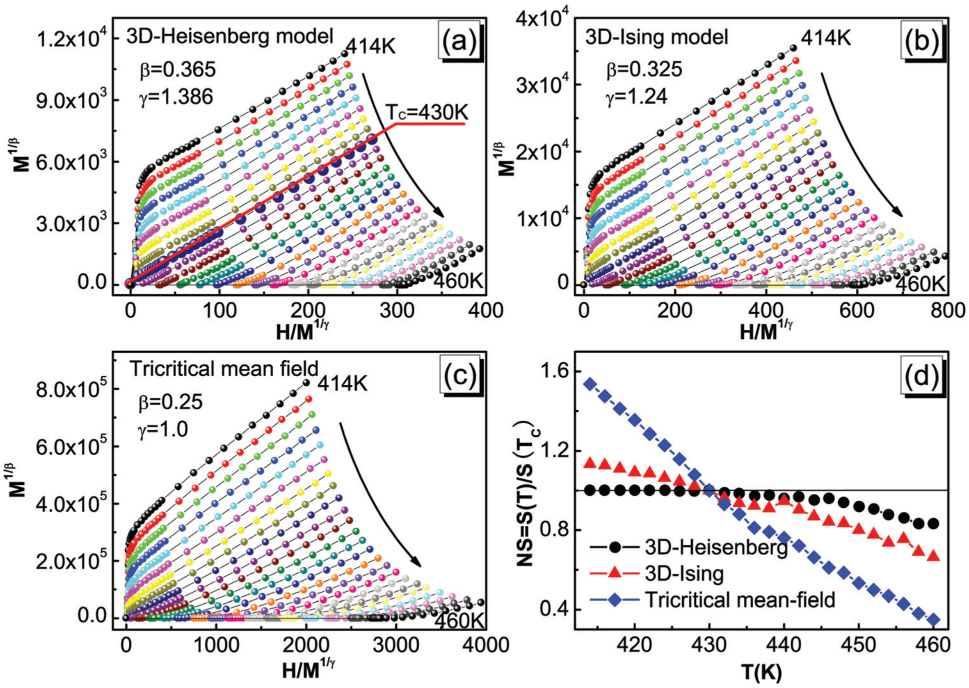

FIG. 2. (Color online) Modified Arrott plot: isotherms of $M^{1/\beta}$ vs $(H/M)^{1/\gamma}$ with (a) 3D-Heisenberg model ($\beta = 0.365$, $\gamma = 1.386$); (b) 3D-Ising model ($\beta = 0.325$, $\gamma = 1.24$); (c) tricritical mean-field model ($\beta = 0.25$, $\gamma = 1.0$); and (d) the normalized slope *NS* [defined as $NS = S(T)/S(T_C)$] as a function of temperature.

$\xi = \xi_0|(T_C - T)/T_C|^{-\nu}$ leads to universal scaling laws for the spontaneous magnetization M_S and initial susceptibility χ_0. The mathematic definitions of the exponents from magnetization can be described as[24,25]

$$M_S(T) = M_0(-\varepsilon)^{\beta}, \varepsilon < 0, T < T_C, \tag{3}$$

$$\chi_0(T)^{-1} = (h_0/M_0)\varepsilon^{\gamma}, \varepsilon > 0, T > T_C, \tag{4}$$

$$M = DH^{1/\delta}, \varepsilon = 0, T = T_C, \tag{5}$$

where ε is the reduced temperature $(T - T_C)/T_C$; M_0/h_0 and D are critical amplitudes; β (associated M_S), γ (associated with χ_0), and δ (associated with T_C) are critical exponents.

In the high magnetic field region, the interplay between charge, lattice, and orbital degrees of freedom are suppressed in a ferromagnet, hence order parameters can be identified with the macroscopic magnetization.[26] As mentioned above, the Landau mean-field theory is not valid for $CuCr_2Se_4$. Therefore, three other kinds of trial exponents of the 3D-Heisenberg model ($\beta = 0.365$, $\gamma = 1.386$), 3D-Ising model ($\beta = 0.325$, $\gamma = 1.24$) and tricritical mean-field ($\beta = 0.25$, $\gamma = 1.0$) are used to make a modified Arrott plot, as given in Figs. 2(a)–2(c). All three models yield quasistraight lines in the high field region. Obviously, the tricritical mean-field model should be excluded first because these straight lines are not parallel to each other. In order to distinguish more clearly, the normalized slop (*NS*), which is defined as $NS = S(T)/S(T_C)$ [where $S(T)$ is the slop of M^2 versus H/M], are plotted in Fig. 2(d). In an ideal model, all values of *NS* should be equal to ′1′ because the modified Arrott plot is a series of parallel straight lines. As for $CuCr_2Se_4$, the *NS* of the 3 D-Heisenberg model is the one close to '1' mostly, and the line of M^2 versus H/M at 430 K just passes through the origin. Thus, the 3D-Heisenberg model is the best one to describe the critical behaviors of $CuCr_2Se_4$.

Based on the modified Arrott plot of Fig. 2(a), the linear extrapolation from high field region to the intercepts with the axes $M^{1/\beta}$ and $(H/M)^{1/\gamma}$ generate the spontaneous magnetization $M_S(T,0)$ and inverse initial susceptibility $\chi_0^{-1}(T,0)$, respectively, which are plotted in Fig. 3(a) as open squares and circles. The experimental data can be fitted according to Eqs. (3) and (4), giving $\beta = 0.371 \pm 0.001$ with $T_C = 430.5 \pm 0.1$ and $\gamma = 1.243 \pm 0.034$ with $T_C = 431.2 \pm 0.4$, respectively. As can be seen that T_C obtained by critical analysis agrees well with that obtained by $M(T)$ curves.

On the other hand, these critical exponents can be also determined more accurately according to the Kouvel–Fisher (KF) method:[27]

$$\frac{M_S(T)}{dM_S(T)/dT} = \frac{T - T_C}{\beta}, \tag{6}$$

$$\frac{\chi_0^{-1}(T)}{d\chi_0^{-1}(T)/dT} = \frac{T - T_C}{\gamma}. \tag{7}$$

According to Eqs. (6) and (7), the $M_S(T)/[dM_S(T)/dT]$ and $\chi_0^{-1}(T)/[d\chi_0^{-1}(T)/dT]$ are as linear functions of temperature, and the slops are $1/\beta$ and $1/\gamma$, respectively. The $M_S(T)/[dM_S(T)/dT]$ and $\chi_0^{-1}(T)/[d\chi_0^{-1}(T)/dT]$ versus T are plotted in Fig. 3(b). The new exponents are obtained as $\beta = 0.372 \pm 0.007$ with $T_C = 430.4 \pm 0.2$ and $\gamma = 1.277 \pm 0.044$ with $T_C = 430.9 \pm 0.5$ according to the KF method. These exponents obtained from the KF method are in agreement with that obtained from the modified Arrott plot.

As confirmation, the critical exponents are tested according to the prediction of the scaling hypothesis. In the critical region, the magnetic equation can be written as[25]

$$M(H, \varepsilon) = \varepsilon^{\beta} f_{\pm}(H/\varepsilon^{\beta+\gamma}), \tag{8}$$

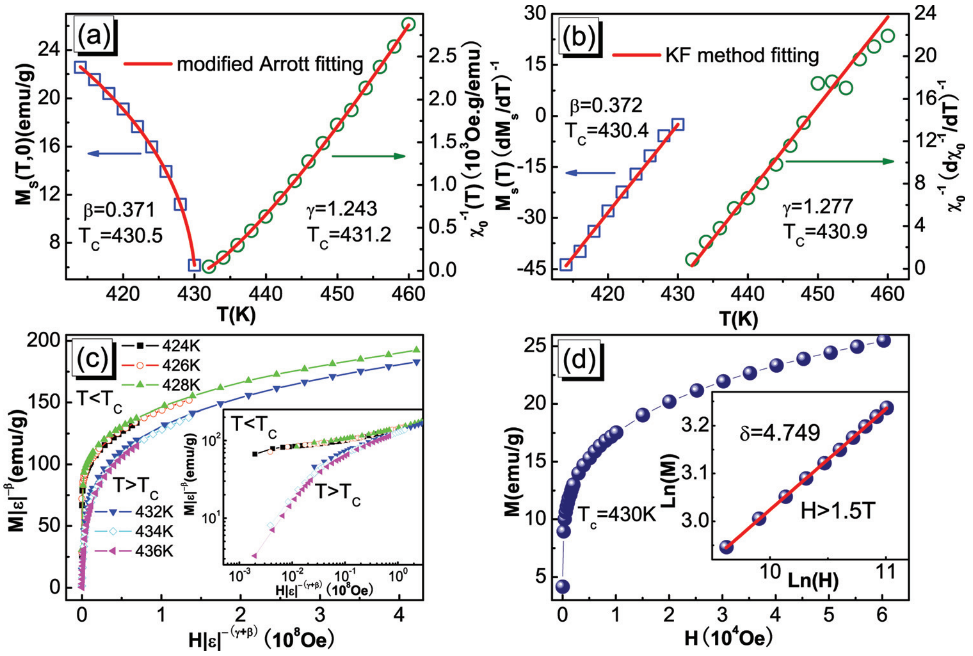

FIG. 3. (Color online) (a)The spontaneous magnetization $M_S(T,0)$ and the inverse initial susceptibility $\chi_0{}^{-1}(T)$ vs T along with the fitting curves (solid curves); (b) KF plot for $M_S(T)$ and $\chi_0{}^{-1}(T)$ (solid lines are fitted); (c) scaling plot below and above T_C using exponents determined from the KF method (only several typical curves are shown); (d) isothermal $M(H)$ at $T_C = 430$ K [the inset shows plots in log-log scale and solid lines are the linear fitting following Eq.(5)].

where $f_\pm$ are regular functions with f_+ for T_C and f_- for $T < T_C$. Equation (8) indicates that $M(H,\varepsilon)\varepsilon^{-\beta}$ versus $H\varepsilon^{-(\beta+\gamma)}$ forms two universal curves for $T > T_C$ and $T < T_C$, respectively. Thus, the isothermal magnetization around T_C are plotted as this prediction of the scaling hypothesis in Fig. 3(c), with the log-log scale in the inset of Fig. 3(c). All experiment data collapses into two different independent curves. This proves that Eq. (8) is obeyed over the entire range of the normalized variables, which indicates the reliability of these obtained critical exponents.

The critical exponents can be also confirmed by the magnetic entropy change ΔS_M, which can be approximated as[28]

$$|\Delta S_M| = \sum \frac{M_i - M_{i+1}}{T_{i+1} - T_i}\Delta H. \tag{9}$$

A phenomenological universal curve for the field dependence of ΔS_M has been proposed as[29–31]

$$\Delta S_M|_{T=T_C} \propto H^n, \tag{10}$$

where $n = 1 + (\beta - 1)/(\beta + \gamma)$. The ΔS_M around T_C are obtained according to Eq. (9). As shown in Fig. 4(a), the ΔS_M presents a unimodal shape and reaches the maximum at T_C when $H \leq 0.8$ T ($\Delta S|_{T=T_C} = \Delta S_M^{\max}$). However, $\Delta S_M(T)$ does not present a unimodal shape when $H > 1$ T. This is because that the ΔS_M in this system originates from the spin-orbit coupling, which can be destroyed by the external magnetic field.[32] It is noted that the $\Delta S_M^{\max}$ is H dependent, as plotted in Fig. 4(b). The $n = 0.629 \pm 0.058$ is obtained according to Eq. (10). On the other hand, through the critical exponents, it can be obtained that $n = 0.619$, which further confirmed that the obtained critical exponents are reliable.

The third critical exponent δ can be determined from the $M(H)$ data at T_C according to Eq. (5). According to the obtained critical exponents above, it can be determined that $T_C = 430$ K. Thus, the isothermal magnetization at $T = 430$ K is given in Fig. 3(d), and the inset of Fig. 3(d) plots the log-log scale. The $\ln(M) - \ln(H)$ relation yields straight line at higher field range ($H > 1.5$ T) with the slop $1/\delta$. Then, it is obtained $\delta = 4.749 \pm 0.016$. According to the statistical

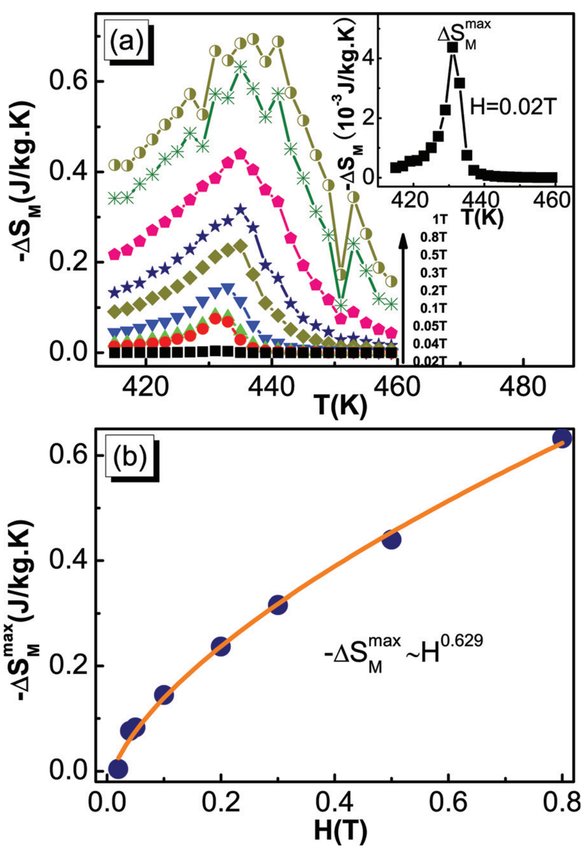

FIG. 4. (Color online) (a) The $\Delta S_M(T)$ for $H \leq 1$ T [the inset shows $\Delta S_M(T)$ for $H = 0.02$ T]; (b) the H dependence of $\Delta S_M^{\max}$ (the solid curve is fitted).

TABLE I. Comparison of critical exponents of $CuCr_2Se_4$ with different theoretical models and other similar systems. MAP = modified Arrott plot; KF = Kouvel–Fisher method; WS = Widom scaling relation.

Material	Ref.	T_C (K)	β	γ	δ
$CuCr_2Se_4$ (MAP)	This work	430.5±0.1	0.371±0.001	1.243±0.034	4.350(WS)
$CuCr_2Se_4$ (KF)	This work	430.4±0.2	0.372±0.007	1.277±0.044	4.433(WS)
$CuCr_2Se_4$ [$M(H)$]	This work	430			4.749±0.016
Tricritical mean-field model	13		0.25	1.0	5.0
Mean-field model	14		0.5	1.0	3.0
3D-Heisenberg model	14		0.365	1.386	4.8
3D-Ising model	14		0.325	1.24	4.82
$CdCr_2Se_4$	18	130	0.337±0.033	1.296±0.109	4.761±0.129
$CdCr_2S_4$	35	85	0.33±0.05	1.44±0.02	5.38±0.01

theory, these three critical exponents agrees with the Widom scaling relation[33]

$$\delta = 1 + \frac{\gamma}{\beta}. \quad (11)$$

Thus, there is $\delta = 4.350$ from the modified Arrott plot in Fig. 3(a) and $\delta = 4.433$ from the KF method in Fig. 3(b), which is slightly smaller than that obtained by the critical isotherm analysis $\ln(M) - \ln(H)$. It is noticed that γ is smaller than the theoretic prediction of the 3D-Heisenberg model, which should be responsible for this deviation.

The obtained critical exponents of $CuCr_2Se_4$, as well as that of different theoretic models and other similar systems, are listed Table I for comparison. It is found that β and δ are close mostly to the 3D-Heisenberg model except that γ is smaller. In fact, γ locates between the 3D-Heisenberg model and the mean-field or tricritical mean-field model. For a homogeneous magnet, the universality class of the magnetic phase transition depends on the exchange interaction $J(r)$. A renormalization group theory analysis suggests the long range attractive interactions decay as[34]

$$J(r) \sim 1/r^{(d+\sigma)}, \quad (12)$$

where d is the spatial dimension, $\sigma > 0$. For three dimension material ($d = 3$), there is relation $J(r) \sim r^{-(3+\sigma)}$ with $3/2 \leq \sigma \leq 2$. When $\sigma = 2$, the Heisenberg exponents ($\beta = 0.365$, $\gamma = 1.386$, and $\delta = 4.8$) are valid for the three dimension isotropic ferromagnet, i.e., $J(r)$ decreases faster than r^{-5}. When $\sigma = 3/2$, the mean-field exponents ($\beta = 0.5$, $\gamma = 1.0$, and $\delta = 3.0$) are valid, which indicates $J(r)$ decreases slower than $r^{-4.5}$. In the present case, β and δ approach the 3D-Heisenberg model mostly, implying that $J(r)$ decreases faster than r^{-5} in the ferromagnetic phase. As for the fact that γ is close to the mean-field model, it indicates that the $J(r)$ decreases slower than $r^{-4.5}$ in the paramagnetic phase, i.e., the correlation length ξ becomes larger than the theoretical prediction of the 3D-Heisenberg model. According to the model proposed by Saha-Dasgupta *et al.*, the ferrimagnetic hybridization between localized electrons of Cr^{3+} ions and delocalized holes of Se 4*p* band results into the half-metallic and ferrimagnetic characteristics in $CuCr_2Se_4$.[4,6,8] Thus, the delocalization of holes produced by Se^{1-} should enlarge the ξ, which should be responsible for the smallness of γ. In other words, the critical exponents obtained here is in agreement with the model proposed by Saha-Dasgupta *et al*.

IV. CONCLUSION

In conclusion, critical properties of $CuCr_2Se_4$ has been comprehensively investigated in the vicinity of critical point T_C based on various techniques including modified Arrott plot, Kouvel-Fisher method, and critical isotherm analysis. Reliable critical exponents of $\beta = 0.372 \pm 0.007$, $\gamma = 1.277 \pm 0.044$, and $\delta = 4.749 \pm 0.016$ around $T_C = 430$ K were obtained. The reliability of these critical exponents were confirmed by the scaling equation, which shows the magnetization-field-temperature (*M-H-T*) data below and above T_C collapse into two different independent curves. These critical exponents were also confirmed by the field dependence of magnetic entropy change relation $\Delta S_M|_{T=T_C} \propto H^n$ with $n = 1 + (\beta - 1)/(\beta + \gamma)$. Results show that the magnetic behaviors of $CuCr_2Se_4$ approach the 3D-Heisenberg model except that γ is smaller than the theoretical value of this model. The delocalized holes of Se 4*p* band should be responsible for the smallness of γ, which supports the model of ferrimagnetic hybridization between localized electrons of Cr^{3+} ions and delocalized holes of Se 4*p* band proposed for $CuCr_2Se_4$.

ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China through Grant No. 11004196, the Knowledge Innovation Program of the Chinese Academy of Sciences through Grant No. 106CS31121 (Hefei institutes of Physical Science, CAS), and the State Key Project of Fundamental Research of China through Grants No. 2007CB925001 and 2010CB923403.

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