Critical behavior and magnetic entropy change of skyrmion host Co$_7$Zn$_8$Mn$_5$

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Abstract

The magnetic properties of a β-Mn-type alloy Co$_7$Zn$_8$Mn$_5$, which is a chiral magnet hosting skyrmion phase, are comprehensively investigated, exhibiting a ferromagnetic transition around 184 K and a spin freezing near 20 K. The generated Rhodes–Wolfarth ratio equals 1.10, which indicates a weak itinerant character of the ferromagnetism in Co$_7$Zn$_8$Mn$_5$. The spin dynamics of the spin freezing agrees with the universal scaling law of critical slowing down with $\tau_0 = 1.7 \times 10^{-5}$ s, $T_g = 20.2$ K, and $z\nu = 3.92$. Critical exponents $\beta = 0.423(1)$ and $\gamma = 1.366(4)$ are deduced by the modified Arrott plot, whereas $\delta = 4.22(2)$ is obtained by a critical isotherm analysis. The validity of the deduced critical exponents is verified by the Widom scaling relation and the scaling hypothesis. The boundary between the first-order and the second-order phase transition is evaluated by a scaling analysis. The magnetic interaction, obtained by a renormalization group theory, decays with distance $r$ as $J(r) \approx r^{-4.9}$, lying between the mean-field model and the 3D Heisenberg model. The analyses on critical behavior could shed new light on the origin of ferromagnetism and topological Hall effect. Moreover, the magnetic entropy change $-\Delta S_M$ exhibits a maximal value around $T_C$, and the peak position gradually raises with increasing fields, eliminating the mean-field model. The $-\Delta S_M^{\max}$ features a power-law behavior with $n > 2/3$, excluding any universal standard models of ferromagnetism. The $-\Delta S_M(T, H)$ plots can be scaled into a universal curve, further verifying the reliability and accuracy of the yielded critical exponents.

1. Introduction

Attracting enormous attentions for emergent electromagnetic responses and possible technological applications in spintronics [1–7], magnetic skyrmions are vortex-like topological spin textures, which are frequently observed in chiral magnets with the Dzyaloshinskii–Moriya interaction (DMI), such as FeGe, MnSi, Co–Zn–Mn alloys, GaV$_4$S$_8$, Cu$_2$OSeO$_3$, and Gd$_2$PdSi$_3$, etc [8–17].

Crystallized in the β-Mn-type crystal structure with space group $P4_3m2$ (No. 213) [inset (i) of figure 1], ternary alloys Co–Zn–Mn are cubic chiral magnets with unique spin helicity, in which the formation of skyrmions lattice (SkL) are confirmed both at and above room temperature, providing a significant step toward applications [17–22]. Recently, electrical manipulation of skyrmions, such as creating, deleting, and driving isolated skyrmions, have been performed in Co–Zn–Mn alloys, producing significance toward the skyrmion-based memory or logic devices [23]. A spin-glass state emerges below the helimagnetic order in Co–Zn–Mn alloys in regions with high Mn concentration [22, 24–26].

Here, we focus on Co$_7$Zn$_8$Mn$_5$, in which a coexistence of helicity and spin-glass state is observed [27]. Remarkably, the intertwined chiral and frustrated magnetism in Co–Zn–Mn alloy generally results in the presence of two equilibrium skyrmion phases, i.e. the A-phase and a disordered skyrmion phase [22, 26]. Furthermore, a giant topological Hall effect (THE) has recently been detected in Co$_7$Zn$_8$Mn$_5$, providing an
Figure 1. (a) Left: magnetization curves as a function of temperature after ZFC and FC processes under an applied magnetic field of 1 kOe. Right: plots of inverse susceptibility $1/\chi$ vs $T$ of the ZFC curve. The linear line represents a fitting by the Curie–Weiss law. Inset: (i) the crystal structure of Co7Zn8Mn5 alloy with a $\beta$-Mn-type crystal structure (space group $P4_1$32). (ii) The plots of derivative magnetization $dM/dT$ vs $T$. (b) $M(H)$ curves at $T = 2$ K and 50 K. Inset: the enlarged $M(H)$ curves.

excellent candidate for investigating the magnetic topological properties and skyrmions in high temperature [27].

A thorough investigation on the magnetic exchange of Co7Zn8Mn5 is of essential importance not only for understanding the underlying basic physics such as merons and nonlinear magnetic order but also for further applications in dictronics based on skyrmion states and other chiral modulations. Especially, the critical behavior analysis on Co7Zn8Mn5, which could shed light on the properties of magnetic interactions, correlation length, the spatial decay of correlation function, and spin dimensionality at criticality [28–31], is still lacking and worthy of a comprehensive characterization. Moreover, the magnetocrystalline anisotropy strongly affects modulated states in cubic chiral magnets [20, 32–34], which takes effects on the critical exponents, deserving a comprehensive investigation.

In this paper, the magnetic properties of Co7Zn8Mn5 are investigated in detail. Based on the measurements on the effective and saturated moments, we yield the Rhodes–Wolfarth ratio (RWR) which equals 1.10, confirming the ferromagnetism in Co7Zn8Mn5 is weak itinerant. The ac susceptibility obeys the universal critical slowing down law. Detailed analyses on critical behavior are carried out. Self-consistent critical exponents are acquired by various methods and verified by scaling analyses. Further analysis reveals a long-range magnetic interaction which decays with distance as $J(r) = r^{-4.9}$. The magnetic entropy change $-\Delta S_M$ is calculated with a maximum of $-\Delta S_M^{\text{max}} = 1.18$ J kg$^{-1}$ K$^{-1}$ under a magnetic field change of 50 kOe. The curves of $-\Delta S_M(T,H)$ can be rescaled into a universal curve, further certifying the accuracy and reliability of the yielded critical exponents.

2. Experimental details

Polycrystalline samples of Co7Zn8Mn5 were fabricated from pure metals of Co, Zn, and Mn with nominal concentrations of 7:8:5. These metals were put in an alumina crucible and subsequently heated in an evacuated quartz tube. Then, the sealed tube was heated to 1273 K, cooled to 1198 K with a rate of 1 K h$^{-1}$, annealed for another 2 days, and quenched into cold water. An ingot of Co7Zn8Mn5 contained some single-crystalline grains (typically up to 2.5 mm) was fabricated by the Bridgman method in an evacuated
quartz tube with cooling from 1298 K to 973 K over 1 week, followed by quenching to cold water [17]. A single-crystalline specimen can be cut out from the resultant product. The phase purity of the sample was checked by powder x-ray diffraction with a β-Mn-type crystal structure. The practical chemical composition was Co$_7$Zn$_8$Mn$_5$, measured by the energy dispersive x-ray spectroscopy, which is close to the nominal composition. The dc and ac magnetic properties of Co$_7$Zn$_8$Mn$_5$ were measured by a magnetic property measurement system (MPMS XL-7, Quantum Design).

3. Results and discussion

3.1. Basic magnetic properties

The plots of zero-field-cooled (ZFC) and field-cooled (FC) magnetization vs $T$ under $H = 1$ kOe are illustrated in the left axis of figure 1(a), in which sharp increases emerge below a transition temperature ($T_C$), implying the presence of a ferromagnetic (FM) order. The first derivative of the ZFC and FC curves ($dM/dT$) vs $T$ are displayed in the inset of figure 1(a) and the temperature of FM-transition is determined to be $T_C \sim 184$ K from the minimum of the $dM/dT$ curves. At temperatures higher $T > 270$ K, a Curie–Weiss behavior is observed for the magnetic susceptibility $\chi = M/H$ (right axis of figure 1(a)). The Weiss $\theta = 231.5$ K and the effective moments $\mu_{\text{eff}} = 10.45~\mu_B$/f.u. were generated by fitted to the Curie–Weiss law, i.e., $\chi = C/(T - \theta)$, where $C$ is a constant related to $\mu_{\text{eff}}$. The magnetization as a function of field $M(H)$ at $T = 2$ K and 50 K is presented in figure 1(b), indicating a typical FM behavior with a saturated moment of $\mu_s = 8.64~\mu_B$/f.u. at low temperature.

With the above generated magnetic parameters of Co$_7$Zn$_8$Mn$_5$, the Rhodes–Wohlfarth ratio (RWR) can be calculated. RWR equals $P_s/P_i$, where $P_s$ equals the saturated moment ($\mu_s$) and $P_i$ is associated with the effective moment, i.e., $P_i(P_i + 2) = \mu_{\text{eff}}^2$ [35, 36]. In a localized FM system RWR is equal to 1, which becomes larger in an itinerant ferromagnetism and/or strong spin fluctuations.

A bifurcation between ZFC and FC curves is detected below around $T = 20$ K (figure 1(a)). Moreover, the coercive force at 2 K is about 1 kOe, while almost no coercive force is observed at 50 K (inset of figure 1(b)). These features remind us of a unusual spin-glass state [24, 25], which is abnormal with the coexistence of a large magnetization. To clarify the unusual spin glass state, temperature-dependent ac magnetic properties were measured with several applied ac driving frequencies ($f$), as displayed in figures 2(a) and (b). Both the real ($m'$) and imaginary ($m''$) part of the ac magnetization exhibit clear dependencies on $f$ with the peak in $m''$ shifts from $T_f = 21.4$ K at $f = 1$ Hz to $T_f = 25.7$ K at $f = 997$ Hz. The frequency-dependent feature is a clear hallmark of spin glass behavior. Note that the magnetization of typical spin glass state should be much weaker, which could due to the existence of a cluster spin glass in Co$_7$Zn$_8$Mn$_5$. The plot of frequency vs $T_f$ can be well characterized by a universal ‘critical slowing down’ model of the spin dynamics [37–39],

$$\tau = \tau_0 \left( \frac{T_f}{T_g} - 1 \right)^{-2z\nu},$$  \hspace{1cm} (1)

where $\tau$ denotes the relaxation time $\tau = 1/f$, $\tau_0$ represents the intrinsic relaxation time, $T_g$ denotes a characteristic temperature for $f \to 0$, and $2z\nu$ represents a critical exponent with the magnitude between 4 and 12. The fitting based on equation (1) is illustrated in inset of figure 2(b), which yields $\tau_0 = 1.7 \times 10^{-3}$ s, $T_g = 20.2$ K, and $z\nu = 3.92$. The achieved value of $\tau_0$ is 7–8 orders of magnitude larger than that of typical single-ion spin-glass systems, in which $\tau_0 \sim 10^{-13}$ s, implying that the relaxation in Co$_7$Zn$_8$Mn$_5$ is dominated by domains or clusters (in which $\tau_0$ could be in the order of $10^{-4}$ s) [40, 41]. The determined value of $z\nu = 3.92$ is close to the theoretically anticipated value for a 3D Ising model with $z\nu = 4$ [37, 42].

The temperature dependence of the zero-field-cooling (ZFC) and field-cooling (FC) dc magnetization at various low fields ($H = 100$, 200, and 500 Oe) is shown in figure 3. Bifurcations between ZFC and FC up to higher temperatures are detected, which could come from the effect from conical/helical or SkL states. More detailed measurements based on small-angle neutron scattering and/or heat capacity with higher resolution are needed to construct a detailed phase diagram.

3.2. Critical behavior

Thoroughly understanding the critical behavior of a material with FM transition is essential, which could provide insight into the underline properties of the spatial decay of the correlation function at criticality, magnetic interactions, the spin dimensionality, the correlation length [28–31]. To offer further comprehension of the FM origin in Co$_7$Zn$_8$Mn$_5$, the isothermal magnetization vs field $M(H)$ close the FM transition temperature around $T_C = 184$ K is measured, and representative plots of $M(H)$ from $T = 165$ K...
Figure 2. (a) The real (in-phase, $m'$) and (b) the imaginary (out-of-phase, $m''$) part of the ac susceptibility as a function of temperature for Co$_7$Zn$_8$Mn$_5$. The amplitude of the ac field is 1 Oe and all the measurements are taken without any applied dc magnetic field. The right panels display the enlarged plots of the related peak region. Inset of (b) is a plot of the relaxation time $\tau = 1/f$ vs $T$ with a fit to the power law, i.e., $\tau = \tau_0(T_p/T_g - 1)^{-\nu}$, generating $\tau_0 = 1.7 \times 10^{-5}$ s, $T_g = 20.2$ K, and $2\nu = 3.92$.

Figure 3. Temperature-dependent ZFC and FC dc magnetization $M(T,H)$ of Co$_7$Zn$_8$Mn$_5$ measured under various indicated magnetic fields. To $T = 201$ K with an interval temperature of $\Delta T = 2$ K are displayed in figure 4(a). The Arrott plots ($M^2$ vs $H/M$) are illustrated in figure 4(b), which are not rigorous straight parallel lines [43], revealing the mean-field theory is not proper to depict the critical phenomenon in Co$_7$Zn$_8$Mn$_5$. The deviation from the mean-field theory is reasonable, considering that the spin fluctuations and Coulomb interactions usually can not be ignored in an itinerant FM material [44, 45], while the mean-field model ignores these impacts. On the basis of the criterion by Banerjee, the order of the FM transition can be decided by the slope of the Arrott plots in high-field region, i.e., a negative slope indicates a first-order transition and the positive one demonstrates a first-order one [46]. The positive slope in figure 2(b) reveals a second-order paramagnetic (PM) to FM transition in Co$_7$Zn$_8$Mn$_5$ in high-field region. The order of the FM transition in low-field region will be investigated afterward.

Since the Arrott plots can not appropriately describe the magnetization in Co$_7$Zn$_8$Mn$_5$, the modified Arrott plots (MAP) are utilized to characterize the $M(H)$ curves. For systems with a second-order FM
Figure 4. (a) Representative measured isothermals $M(H)$ from $T = 165$ to $201$ K with a temperature interval of $\Delta T = 2$ K. (b) Plots of $M^2$ vs $H/M$ (the Arrott plot).

transition, the spontaneous magnetization ($M_s$) below $T_C$, the $M(H)$ curve at $T_C$, and the initial magnetic susceptibility ($\chi^{-1}$) above $T_C$ are depicted by various critical exponents $\beta$, $\delta$, and $\gamma$, respectively [47, 48],

$$M_s(T) \propto |\varepsilon|^\beta, \quad \varepsilon < 0, \quad T < T_C,$$

$$M \propto H^{1/\delta}, \quad \varepsilon = 0, \quad T = T_C,$$

$$\chi^{-1}(T) \propto |\varepsilon|^\gamma, \quad \varepsilon > 0, \quad T > T_C,$$

in which $\varepsilon = (T - T_C)/T_C$ represents a scaled temperature. Several universal theoretical models, i.e., the 3D Ising model with $\beta = 0.325$ and $\gamma = 1.24$, 3D Heisenberg model with $\beta = 0.365$ and $\gamma = 1.386$, 3D XY model with $\beta = 0.345$ and $\gamma = 1.316$, and tricritical mean-field model with $\beta = 0.25$ and $\gamma = 1.0$ are utilized to figure out the MAP, as shown in figures 5(a)–(d). A proper model should lead to various parallel lines with the same slope in the high-field region. Comparing the normalized slope $NS = S(T)/S(T_C)$ with the ideal value 1 could enable us to discover the best model. The $NS$ of all the universal models exhibit considerable departure from 1 (figure 6(a)), revealing that the above universal models are not applicable in Co$_7$Zn$_8$Mn$_5$.

To achieve proper critical exponents, an iterative way is utilized to determine the MAP [49–52], which successfully generates a series of parallel lines in the high-field region (figure 6(b)). Linearly extrapolated $M_s$ and $\chi^{-1}$ as a function of temperature are displayed in figure 7(a). Based on equations (2) and (4), the solid fits generate $\beta = 0.423(1)$, with $T_C = 183.6(1)$ K, and $\gamma = 1.366(4)$, with $T_C = 183.7(1)$ K (figure 7(a)). Furthermore, a Kouvel–Fisher (KF) method can also be exploited to acquire critical exponents better precision [53],

$$M_s(T)/(dM_s(T)/dT) = (T - T_C)/\beta,$$

$$\chi^{-1}(T)/(d\chi^{-1}(T)/dT) = (T - T_C)/\gamma.$$

As illustrated in figure 7(b), the determined values of critical exponents by linear fits are $\beta = 0.422(3)$, with $T_C = 183.5(4)$ K, and $\gamma = 1.365(4)$, with $T_C = 183.7(5)$ K, respectively, consistent with those values yielded by the iterative MAPs, revealing that the obtained exponents are reliable and intrinsic.
Figure 5. The MAP, i.e., $M^{1/\beta}$ vs $(H/M)^{1/\gamma}$ with exponents belonging a (a) 3D Ising model, (b) 3D Heisenberg model, (c) 3D XY model, and (d) tricritical mean-field model.

On the basis of equation (3), the plot of $\log_{10}(M)$ vs $\log_{10}(H)$ at $T_C$ should result in linear line in the high-field region with the fitted slope equals to $1/\delta$ (the inset of figure 7(a)), where $\delta = 4.22(2)$ is obtained by the linear fit. Based on the Widom’s law [54],

$$\delta = 1 + \gamma/\beta,$$

the exponents $\delta = 4.23(1)$ and $\delta = 4.23(2)$ can be calculated by exploiting the critical exponents $\beta$ and $\gamma$ from the MAP and KF methods, respectively. Both values of $\delta$ are close to that from the critical isothermal analysis at $T_C$, indicating self-consistency of the yielded critical exponents.

The reliability of the achieved critical exponents can be further checked by scaling analyses. The scaling plot of FM systems in the asymptotic region is the following equation [54],

$$M(H, \varepsilon) = \varepsilon^{\beta} f'_\pm (H/\varepsilon^{\beta+\gamma}),$$

where $f'_-$ for $T < T_C$ and $f'_+$ for $T > T_C$, respectively, are normal functions for scaling. With appropriately determined values of $\beta$, $\gamma$, $\delta$, and $T_C$, the plots of scaled magnetization ($m = M|\varepsilon|^{-\beta}$) vs scaled field ($h = H|\varepsilon|^{-(\beta+\gamma)}$) should collapse onto two branches below and above $T_C$, respectively. As presented in figure 8, all the data of $m$ vs $h$ in the high-field region fall into two universal branches below and above $T_C$. Moreover, the scaling function can exhibit another composition [47],

$$\frac{M}{H^{1/\gamma}} = g\left(\frac{\varepsilon}{H^{1/\gamma}}\right),$$

where $g$ represent a scaling function. With a proper scaling by equation (9), all the curves would collapse onto a universal one. As expected, the plots of $\frac{MH^{-1/\beta}}{\varepsilon H^{-1/\gamma}}$ in the high-field region fall into a single universal curve (inset of figure 8). The well scaling behaviors reveals that the obtained exponents are trustworthy and consistent with the above hypothesis of scaling. The defects and grain boundary usually exist in polycrystalline samples. However, the correlation length $\xi$ depends on the temperature change as,

$$\xi = \xi_0 |\varepsilon|^{-\nu},$$

where $\varepsilon = (T - T_C)/T_C$. Thus, when the temperature is close to $T_C$, the correlation length will be very large, and the effects come from sample details, such as the disorders, grain boundaries, defects, or vacancies, should be small compared with so large a scale of the correlation length [55]. In other words, the correlation length is insensitive to the sample details when temperature is close to $T_C$ [56, 57].
Figure 6. (a) $\text{NS}_S = S(T)/S(T_c)$ as a function of temperature for various universal theoretical models. (b) The generated MAP of $M^{1/\alpha}$ vs $(H/M)^{1/\gamma}$, with $\beta = 0.423(1)$ and $\gamma = 1.366(4)$.

Additionally, a comprehensive measurement on the low-field region is executed, and the experimental data below $T_c$ in the low-field region cannot be scaled into one universal curve, as illustrated in figure 9. The plots of $m^2 vs h/m$ with a log10-log10 scale exhibit cusps in the low-field region. As we know, in the low-field region of Co–Zn–Mn alloys exists conical/helical or SkL states [18, 22], where usually presents first-order phase transitions under relatively low external magnetic fields [58]. The low-field first-order transition could be tuned to a second-order one with larger applied magnetic fields [59, 60]. We argue that it is due to the presence of the first-order phase transition below $H_C$, the scaling equation becomes invalid. Denoted by arrow in figure 9, the magnetic field $H_C$ usually lies at the boundary of the first-order phase transition and the second-order one. At $T_C$ the $H_C$ is about 180 Oe, and the order of magnitude is consistent with previous reported values where the SkL phase presents [17, 22, 27]. In a recent study, it has been suggested [58] that the renormalization proposed by Brazovskii [61] could be utilised to characterize the nature of the weak first-order the PM to helimagnetic transition near zero magnetic field in skyrmion host MnSi [62, 63]. According to this proposal, the renormalization arises from the DMI which changes the nature of the spin fluctuations near $T_C$, and causes the system avoiding the second-order phase transition expected by the mean-field model. By the way, the existence of first-order FM transitions in low-field region makes it difficult to achieve the critical exponents by heat capacity and/or thermal expansion [60, 64].

Recently, magnetocrystalline anisotropy has been argued to make an important effect in the formation of a tilted conical state and a low-temperature SkL state in skyrmion hosts Cu2OSeO3 and Co–Zn–Mn alloys [32, 65]. To distinguish the importance of the anisotropy in Co7Zn8Mn5, we evaluated the magnetocrystalline anisotropy $K_u$, which is associated with the saturated magnetic field $H_i$ and the saturated magnetization $M_s$ under the field $H//ab$ plane, i.e., $2K_u/M_s = \mu_0 H_i$, where $\mu_0$ represents the vacuum permeability [66]. The yielded $K_u$, displayed in the left axis of figure 10, decreases with an increasing temperature, exhibiting a temperature dependency, which is common and could be derived from local spin-clusters fluctuating and activated by thermal energy [67, 68]. The determined value of $K_u$ equals 185.1 kJ m$^{-3}$ at $T = 130$ K in Co7Zn8Mn5. On the basis of a classical theory, $\langle K_u \rangle \propto M_s^{p(n+1)/2}$, where $M_s$
Figure 7. (a) Plots of spontaneous magnetization $M_s$ and inverse initial susceptibility $1/\chi_0$ vs $T$ with solid fitting curves for Co$_7$Zn$_8$Mn$_5$. Inset: the $M(H)$ curve at $T_C = 184$ K in log$_{10}$–log$_{10}$ scale with linear fits. (b) Plots of $M_s/(dM_s/dT)$ and $\chi_0^{-1}/(d\chi_0^{-1}/dT)$ vs $T$ (the KF plots) with linear fits.

Figure 8. Scaling plots of scaled magnetization $m = M|\varepsilon|^{-\beta}$ vs scaled field $h = H|\varepsilon|^{-\gamma}$ with exponents $\beta = 0.423$, $\gamma = 1.366$, and $T_C = 184$ K. Inset: another scaling plots of $M|H^{\beta}\varepsilon^{-\gamma}|$ vs $H^{1/\gamma}$.

denotes the saturated magnetization below $T_C$ and $\langle K_u^{(\mu)} \rangle$ represents an expectation value of the magnetocrystalline anisotropy constant [67, 68], in the case of an uniaxial anisotropy $n = 2$ and in a cubic one $n = 4$, resulting in exponents of 3 and 10, respectively. To test whether the above model can be utilized to characterize the temperature dependence of $K_u$ in Co$_7$Zn$_8$Mn$_5$, we display in the right axis of figure 10 the evaluated $M_s/M_s(130 \text{K})$, $[M_s/M_s(130 \text{K})]^3$, and $[M_s/M_s(130 \text{K})]^{10}$ as a function of $T$. The comparison clearly indicates that the magnetocrystalline anisotropy in Co$_7$Zn$_8$Mn$_5$ cannot be characterized by the simple classical model with an uniaxial anisotropy or a cubic one, which is consistent with a previous report that the anisotropy in Co$_7$Zn$_8$Mn$_5$ sharply varies upon changing composition and/or temperature [65], and could be related to temperature- and composition-induced deformations of skyrmions, variations of the
helical modulation vectors, rearrangements of the structure of SkLs, and the enhancement of Gilbert damping [65].

In table 1, we display the exponents yielded by various methods and those from several universal theoretical models. The exponents of various related skyrmion materials are also shown. The critical exponent $\beta$ of Co$_7$Zn$_8$Mn$_5$ approaches that of mean-field model, while $\gamma$ is close to that of the 3D Heisenberg model. The magnitude of $\beta$ for two-dimensional magnets should be in in the range of $0.1 \leq \beta \leq 0.25$ [74]. Clearly, the critical exponents of Co$_7$Zn$_8$Mn$_5$ reveal a 3D critical behavior, consistent with its three-dimensional crystal structure. Then we compare the exponents with the other skyrmion hosts. The critical exponents of Néel-type skyrmion host GaV$_4$S$_8$ are respond to the tricritical mean-field model [70]. The critical behavior of the Bloch-type skyrmion hosts Fe$_{0.8}$Co$_{0.2}$Si and FeGe can be described by the 3D Heisenberg theory [71, 72], while MnSi approaches the tricritical mean-field model [59]. For Cu$_2$OSeO$_3$, its critical exponents are characterized by the 3D Heisenberg theory above $T_C$ [73, 75]. The effect of anisotropy plays an vital role in Co–Zn–Mn alloys [32, 65], the critical exponents will be facilitated by DMI, anisotropy below $T_C$, symmetric exchange interaction. Similar anisotropic effect is also detected in cubic chiral skyrmion host Cu$_2$OSeO$_3$, in which the magnetocrystalline anisotropy and anisotropy due to spin–orbit coupling increases the DMI, leading to a 3D Ising-type interaction below $T_C$ [75]. For comparison, the exponents of Fe$_{0.8}$Co$_{0.2}$Si are close to the 3D Heisenberg model [72], exhibiting an isotropic short-range magnetic coupling. In Co$_7$Zn$_8$Mn$_5$, the cubic anisotropy strongly affects the modulated states in chiral magnets, e.g., determines the favored $q$-vector directions, changes the stability range of the SkL state and can also introduces new modulated states [20, 32–34]. The anisotropic magnetic exchange interaction could lead to the discrepancy of the critical exponents to a 3D Heisenberg model.
Table 1. Comparison of the yielded critical exponents with various universal theoretical models and several skyrmion hosts. The MAP, KFP, CI, and cal represent the MAP, the KF plot, the critical isotherm, and calculated, respectively.

| Composition          | Reference | Technique | $T_C$    | $\beta$ | $\gamma$ | $\delta$ | $n$ | $m$ |
|----------------------|-----------|-----------|----------|---------|----------|----------|-----|-----|
| This work            | MAP       | 183.6(1)  | 0.423(1) | 1.366(4) | 4.23(2)$^{\text{uf}}$ |          |     |     |
| This work            | KFP       | 183.7(1)  | 0.422(3) | 1.365(4) | 4.23(2)$^{\text{uf}}$ |          |     |     |
| This work            | CI        | 184       |          |         |          | 4.22(2)  |     |     |
| This work - $\Delta S_{\text{calf}}$ | RCP   | 184       |          |         |          | 0.782(3) |     |     |
| Co$_2$Zn$_9$Mn$_3$   | This work | 184       |          |         |          |          |     |     |
| 2D Ising             | [69]      | Theory    | 0.125    | 1.75    | 15       | 0.533    | 1.06 |
| Mean field           | [43]      | Theory    | 0.5      | 1.0     | 3.0      | 0.667    | 1.333|
| 3D Ising             | [43]      | Theory    | 0.325    | 1.24    | 4.82     | 0.569    | 1.207|
| 3D Heisenberg        | [43]      | Theory    | 0.365    | 1.386   | 4.8      | 0.637    | 1.208|
| 3D XY                | [43]      | Theory    | 0.345    | 1.316   | 4.81     | 0.606    | 1.208|
| Tricritical mean field| [46]    | Theory    | 0.25     | 1.0     | 5        | 0.4      | 1.20 |
| Ga$_4$V$_8$S $_8$    | [70]      | MAP       | 12.0     | 0.220(24)| 0.999(5) | 5.13(11) |     |     |
| FeGe                 | [71]      | MAP       | 283      | 0.336(4) | 1.352(3) | 5.267(1) |     |     |
| Fe$_{48}$Co$_{28}$Si | [72]      | Hall      | 36.0(5)  | 0.371(1) | 1.382(1) | 4.78(1)  |     |     |
| MnSi                 | [59]      | MAP       | 30.5     | 0.242(6) | 0.915(3) | 4.734(6) |     |     |
| Cu$_2$OSe$_3$        | [73]      | AC        | 58.3(1)  | 0.37(1)  | 1.44(4)  | 4.9(1)   |     |     |

below $T_C$. As $\beta$–Mn-type cubic chiral magnets, Co–Zn–Mn alloys with a chiral space group are DMI-based SKL, in which skyrmions with unique spin helicity form both at and above room temperature [17, 25]. Previous systematical investigations on DMI demonstrated that the DMI in Co–Zn–Mn alloys is controlled by the change in band filling and that in principle the sign reversal of DMI universally occurs in metallic chiral magnets with easily tunable band filling [76].

Then, we make a discussion on the origin as well as the range of interactions of Co$_7$Zn$_8$Mn$_5$ based on a renormalization group theory, which reveals a long-range interaction with attraction would decay with distance $r$ as $J(r) \propto r^{-(d+\sigma)}$, where $d$ represents the spatial dimension and $\sigma$ is associated with the range of interaction [77]. For a long-range interaction, $\sigma$ could be achieved by $[49, 77, 78]$,

$$
\gamma = 1 + \frac{4(n + 2)}{d(n + 8)} \Delta \sigma + \frac{8(n + 2)(n - 4)}{d^2(n + 8)^2} \times \left[ 1 + \frac{2G(\frac{d}{2})}{(n + 4)(n + 4)} \right] \Delta \sigma^2,
$$

where $d$ denotes the spatial dimension, $n$ represents the spin dimensionality, $G(\frac{d}{2}) = 3 - \frac{1}{2}(\frac{d}{2})^2$, and $\Delta \sigma = \sigma - d/2$. For a system with three-dimensionality, $d$ should be equal to 3, and the relation of decay becomes $J(r) \propto r^{-(3+\sigma)}$. When $\sigma = 3/2$, the mean-field model takes effect and $J(r)$ decays with distance $r$ slower than $r^{-4.5}$. When $\sigma = 2$, the Heisenberg model is valid for a 3D isotropic FM system and $J(r)$ decays faster than $r^{-5}$. In Co$_7$Zn$_8$Mn$_5$, the magnetic exchange is determined to decay with distance $r$ as $J(r) \propto r^{-4.9}$, lying between the 3D Heisenberg model and the mean-field mode with a long-range interaction $[49, 77, 78]$.

3.3. Magnetic entropy change

On the basis of the thermodynamical theory and the Maxwell relations, the magnetic entropy change can be evaluated as,

$$
\Delta S_M(T, H) = \int_0^H \left( \frac{\partial M}{\partial T} \right)_H dH.
$$

 Practically, $\Delta S_M(T, H)$ can be evaluated as,

$$
\Delta S_M = \frac{\int_0^H M(T_i, H) dH - \int_0^H M(T_{i+1}, H) dH}{T_i - T_{i+1}},
$$

when magnetization $M(H)$ are measured at relatively small intervals of magnetic field and temperature. The calculated $-\Delta S_M(T, H)$ as a function of temperature under various fields is illustrated in figure 11. A peak presents around $T_C$ in all the plots of $-\Delta S_M(T, H)$ vs $T$, and these peaks expand asymmetrically on both sides with an increasing applied magnetic field, exhibiting typical behavior of FM systems. In addition, the position of the peaks gradually transfer to higher temperatures with an increasing of applied magnetic fields, which eliminates the mean-field model $[79–81]$, consistent with the above analyses on critical behavior. $-\Delta S_M(T, H)$ reaches to a maximal value of 1.18 J kg$^{-1}$ K$^{-1}$ at $\mu_0 H = 5$ T, the value is comparable with that of typical systems with a second-order FM transition $[51, 82, 83]$.

Except for $-\Delta S_M$, another crucial parameter to characterize the potential importance of the magnetocaloric effect of systems is the RCP $[84]$, which equals $-\Delta S_M^{\text{max}} \times \delta_{\text{FWHM}}$, where $-\Delta S_M^{\text{max}}$
Figure 11. Temperature dependence of the calculated magnetic entropy change for Co$_7$Zn$_8$Mn$_5$ under various magnetic fields.

Figure 12. The field-dependent $\Delta S_M^{\text{max}}$ (left) and RCP (right). Inset (i): plots of $n$ vs $T$ under various fields.

represents the maximal value of the $-\Delta S_M$, $\delta\nu_{\text{FHM}}$ is the full width at half maximal value of $-\Delta S_M(T)$. Plots of $-\Delta S_M^{\text{max}}$ and RCP vs $T$ can be well depicted by the following relations of power law,

$$-\Delta S_M^{\text{max}} \propto H^n,$$

$$\text{RCP} \propto H^m,$$

where $n$ and $m$ are associated with the above exponents as follows [85, 86],

$$n(T_C) = 1 + (\beta - 1)/(\beta + \gamma),$$

$$m = 1 + 1/\delta.$$

As illustrated in the left and right axes of figure 12, $n = 0.782(3)$ and $m = 1.068(2)$ are fitted based on equations (13) and (14), respectively. Any values of $n$ larger than 2/3 cannot be understood with the universal classes of models for FM system (XY, Heisenberg, mean field, and Ising) [25], which is consistent with the above analyses on critical behavior. Elevated values of $n$ are generally exhibited for FM systems with bulk magnetic glasses [25, 87], in which magnetic interactions are disordered. Inset of figure 12 displays the plots of $n(T)$, which is yielded by $n = \frac{\ln|\Delta S_M|}{\ln(H)}$. The behavior of $n$ agrees well with the universal law of FM systems [88], i.e., at low temperature, far below $T_C$, $n$ is close to 1; at high temperature, far above $T_C$, $n$ approaches 2 owing to the Curie–Weiss law; at $T = T_C$, $n$ reaches a minimum.

In addition, a scaling analysis can be performed for systems with a second-order FM transition [89]. In this scaling process the scaled magnetic entropy change, i.e., $\Delta S_M/\Delta S_M^{\text{max}}$, calculated for various fields, is scaled to a reduced temperature $t$. 
Figure 13. (a) The scaled magnetic entropy change $\Delta S_M/\Delta S_{\text{max}}$ as a function of the reduced temperature $t$. Inset: $T_{r1}$ and $T_{r2}$ as a function of field. (b) Scaling analysis of $\Delta S_M(T)$ using the generated critical exponents.

\begin{align*}
t_- &= (T_{\text{peak}} - T)/(T_{r1} - T_{\text{peak}}), \quad T \leq T_{\text{peak}}, \\
t_+ &= (T - T_{\text{peak}})/(T_{r2} - T_{\text{peak}}), \quad T > T_{\text{peak}},
\end{align*}

in which $T_{r1}$ and $T_{r2}$, which are two reference temperatures of the full width at half maximum $\Delta S_M(T_{r1}, T_{r2}) = \frac{1}{2} \Delta S_{\text{max}}$, are exhibited in inset of figure 13(a). In this scaling process, $T_C$ is not a good parameter due to its field dependence, whereas the peak temperature ($T_{\text{peak}}$) serves the purpose. Exploiting the above scaling method, the normalized $\Delta S_M/\Delta S_{\text{max}}$ falls into a universal curve around $T_{\text{peak}}$ (figure 13(a)), which further verifies FM transition in Co$_7$Zn$_8$Mn$_5$ under high field is of a second order. The low temperature deviation should be due to the spin-glass behavior and/or the effects from SkL state. Additionally, the scaling analysis of systems with a second-order FM transition could also be implemented by the scaling function,

\[ -\Delta S_M \propto H^n h \left( \frac{\varepsilon}{H^{1/(\beta+\gamma)}} \right), \]

where $n$, $\beta$, and $\gamma$ denote critical exponents and $h$ represents a regular function [90]. With properly chosen exponents, $\Delta S_M(T)$ vs $H^{1/(\beta+\gamma)}$ curves should be renormalized to a universal curve, which is indeed observed around $T_{\text{peak}}$ (figure 13(b)). The well-renormalized $-\Delta S_M(T, H)$ curves around the FM transition verifies the validity and reliability of the generated critical exponents. A disconvergence of the $\Delta S_M(T, H)$ data at low temperature is also detected, which should be due to the presence of the spin freezing and/or the effects from SkL state.

For the investigation and modulation of skyrmion physics and related topological properties in skyrmion hosts, a systematic investigation of the critical exponents and associated magnetic interactions is of significant importance [47, 55, 72]. In Cu$_2$OSeO$_3$ and other SkLs, one can discover first-order transitions and narrow dips just below the transition by a critical scaling analysis [59, 73], which are hallmarks of the formation of SkL. In MnSi, investigations of the critical behavior and polarized neutron scattering lead to the proposal of the presence of a ‘skyrmion-liquid’ phase in a narrow-temperature range [62, 64]. Furthermore, critical behavior analyses can yield the spin–orbital coupling and magnetic interaction, which exhibits essential impact on the presence and development of nontrivial topological states and on the measured THE. In PrAlGe, the FM order gives rise to the band splitting, which induces breakdown of time-reversal symmetry by the movement of Weyl nodes shift, bringing about a large anomalous Hall effect.
[91, 92]. In CeAlGe, it is certificated that several magnetic incommensurate phases exist, which implies close correlations between topologically nontrivial states and magnetism [93]. Thus, the detailed investigation of the FM properties in Co7Zn8Mn5 is of substantial importance for comprehending the interplay between the magnetic properties and skyrmion-physics and could help offering explanations to the THE in Co7Zn8Mn5.

4. Conclusions

In summary, the magnetic properties of skyrmion host Co7Zn8Mn5 are systematically investigated. The generated RWR reveals a weak itinerant ferromagnetism at \( T_C = 184 \) K. The ac susceptibility measurements on the spin-glass state at \( T_g = 20.2 \) K confirms the relaxation in Co7Zn8Mn5 is dominated by clusters and/or domains rather than single ion effects. Critical exponents \( \beta = 0.423(1), \gamma = 1.366(4), \) and \( \delta = 4.22(2) \) are yielded by analyses on critical behavior. The reliability and self-consistency of the achieved exponents are verified by the scaling analyses. The magnetic interaction in Co7Zn8Mn5 is of a long range and the exchange interaction \( J(r) \) decays with distance \( r \) as \( J(r) = r^{-4.9} \). Moreover, the scaling analysis on magnetic entropy change \( -\Delta S_M(T, H) \) further verifies that the yield critical exponents are accurate and intrinsic. Considering the existence of two equilibrium skyrmion phases and a giant THE observed in Co7Zn8Mn5, a comprehensive investigation on its magnetic properties is valuable. Our work provides valuable information for future study.

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Data availability statement

The data are available from the corresponding author on reasonable request.

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