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# 3D-Heisenberg magnetic coupling in the skyrmion system $Fe_{1.5-x}Co_xRh_{0.5}Mo_3N$ ($x = 0.5, 1.0, 1.2$)

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ABSTRACT

Critical behaviors of $Fe_{1.5-x}Co_xRh_{0.5}Mo_3N$ ($x = 0.5, 1.0, 1.2$) are investigated, which are well known as a new family hosting skyrmion phase. The critical exponents $\beta$, $\gamma$, and $\delta$, which correspond to the magnetization, are obtained by the modified Arrott plot and critical isothermal analysis. The reliability of these critical exponents are testified by the Widom scaling law and scaling equation. The critical exponents of $Fe_{1.5-x}Co_xRh_{0.5}Mo_3N$ ($x = 0.5, 1.0, 1.2$) are close to the universality class of 3D-Heisenberg model, which suggests that the magnetic coupling in this system is of an isotropic short-range type.



## 1. Introduction

The skyrmion phase has attracted considerable attention due to the topological property [1,2], nanometric size [3,4], and current-driven motion [5–8], which has great potential application in the next generation spintronic storage [9,10]. However, the narrow windows of temperature and magnetic field block its application. Therefore, the exploration and investigation of new materials hosing skyrmion phase are significant not only in the fundamental physics but also in applications [11,12]. Recently, skyrmion phase is found in the modulated molybdenum nitrides $A_2Mo_3N$ system, a new system different from conventional ones [13]. The cubic $A_2Mo_3N$ (A = Fe, Co, and Rh) has the filled $\beta$-manganese structured cell, the space group of which belongs to $P4_132$ [14–16]. The atoms on A-sites lie on the 8*c* positions forming a single (10, 3)-*a* network [17]. This network is filled by corner-shared $Mo_6N$ octahedra. The parent compounds $Fe_2Mo_3N$, $Co_2Mo_3N$, and $Rh_2Mo_3N$ exhibit ferromagnetic, antiferromagnetic, and superconductive state, respectively [13,18].

The antisymmetric structure in $A_2Mo_3N$ results in the lack of inversion symmetry. Therefore, Dzyaloshinskii-Moriya (DM) interaction is predicted in $A_2Mo_3N$, which is usually an important requirement in forming a skyrmion phase [19,20]. Study has shown that the doping on A-sites can effectively tune the strength of the spin-orbital coupling, which in turn modulates the DM interaction and ferromagnetic coupling [16]. It has been demonstrated that $Fe_{1.5}Rh_{0.5}Mo_3N$ exhibits a ferromagnetic transition at ~110 K, the highest phase transition temperature in $Fe_{2-x}Rh_xMo_3N$ system [16,21]. By proper modulation of A-sites, the skyrmion phase has been subsequently observed in the $Fe_{1.5-x}Co_xRh_{0.5}Mo_3N$ system [13]. It has been demonstrated that Fe, Co, and Rh atoms contribute significantly to the energy-band near the Fermi surface and are responsible for magnetism rather than Mo atoms [13].

Although the skyrmion phase in molybdenum nitrides $A_2Mo_3N$ system has been investigated, its magnetic coupling still remains open. In this work, we reveal the magnetic coupling of $Fe_{1.5-x}Co_xRh_{0.5}Mo_3N$ ($x = 0.5, 1.0, 1.2$) through the investigation of the critical behaviors. The critical exponents $\beta$, $\gamma$, and $\delta$, which correspond to the magnetization, are obtained by the modified Arrott plot and critical isothermal analysis. In addition, the reliability of these critical exponents are testified by the Widom scaling law and

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scaling equation. The critical exponents of $Fe_{1.5-x}Co_xRh_{0.5}Mo_3N$ ($x = 0.5, 1.0, 1.2$) belong to the universality class of 3D-Heisenberg model, which suggests an isotropic short-range magnetic coupling in this system.

## 2. Experimental methods

Polycrystalline samples of $Fe_{1.5-x}Co_xRh_{0.5}Mo_3N$ ($x = 0.5, 1.0, 1.2$) were synthesized by the reductive nitridation of a binary oxides mixture [14–16]. The detailed sample preparation method and physical properties were described and checked elsewhere [13]. The magnetization was measured using a Quantum Design Vibrating Sample Magnetometer (SQUID-VSM). The no-overshoot mode was applied to ensure a precise magnetic field. The magnetic field was relaxed for two minutes before data collection. For the measurement of initial isothermal magnetization, the sample was firstly heated adequately above the Curie temperature $T_C$ for ten minutes, then cooled to the target temperature under zero magnetic field.

## 3. Results and discussion

Fig. 1 (a), (b), and (c) give the temperature dependence of magnetization [$M(T)$] (left axis) and the derivative curves ($dM/dT$) (right axis) for $Fe_{1.5-x}Co_xRh_{0.5}Mo_3N$ ($x = 0.5, 1.0, 1.2$). The $M(T)$ curves are measured under sequences of zero-field-cooling (ZFC) and field-cooling (FC). All $M(T)$ curves exhibit magnetic phase transitions with the decrease of temperature. A bifurcation between ZFC and FC appears below the phase transition temperature for each $M(T)$ curve, which indicates a ferromagnetic cluster behavior. The phase transition temperatures $T_C$ are determined by the $dM/dT$ curves, where $T_C$ are determined as ~120.5 K, ~82.5 K, and ~34.5 K for $x = 0.5, 1.0$ and 1.2, respectively. It can be seen that $T_C$ decreases with the increase of Co-content. In addition, the phase transition for $x = 0.5$ is much sharper than that for $x = 1.2$. These results indicate that the phase transition becomes weaker with the increase of Co.

Fig. 2 depicts the isothermal magnetization as a function of field [$M(H)$] at 4 K for $Fe_{1.5-x}Co_xRh_{0.5}Mo_3N$ ($x = 0.5, 1.0, 1.2$). All $M(H)$ curves show magnetic ordering behaviors. The inset gives the saturation magnetization ($M_S$) vs. the Co-content $x$. It shows that $M_S$ decreases with the increase of $x$, which suggests the weakening of the magnetization by the doping of Co. The results form the $M(H)$ curves are in agreement with the indication from $M(T)$ curves.

In order to uncover the magnetic interaction in $Fe_{1.5-x}Co_xRh_{0.5}Mo_3N$ system, the critical behavior should be investigated. Fig. 3 (a), (b), and (c) show the initial isothermal magnetization [$M(H)$] around $T_C$ for $Fe_{1.5-x}Co_xRh_{0.5}Mo_3N$ ($x = 0.5, 1.0, 1.2$), respectively. The measurement of the initial $M(H)$ was limited within the asymptotic critical temperature region, *i. e.* $|(T - T_C)/T_C| < 0.1$. Generally, the Arrott plot can roughly reveal the order and critical temperature of the magnetic phase transition. The Arrott plot of $M^2$ vs. $H/M$ curves are shown in Fig. 3 (d), (e), and (f) for $x = 0.5$, $x = 1.0$, and $x = 1.2$, respectively. According to Banerjee's criterion, a negative slope suggests a first-order transition while a positive one implies a second-order one [22]. Therefore, the positive slopes of the $M^2$ vs. $H/M$ curves for $Fe_{1.5-x}Co_xRh_{0.5}Mo_3N$ indicate that the phase transitions in all these samples are of second-order types. More generally, the initial $M(H)$ curves in the asymptotic critical temperature region follow the Arrott-Noakes equation of state [23]:

$$(H/M)^{1/\gamma} = (T - T_C)/T_C + (M/M_1)^{1/\beta} \quad (1)$$

where $M_1$ is a constant, $\beta$ and $\gamma$ are critical exponents. According to

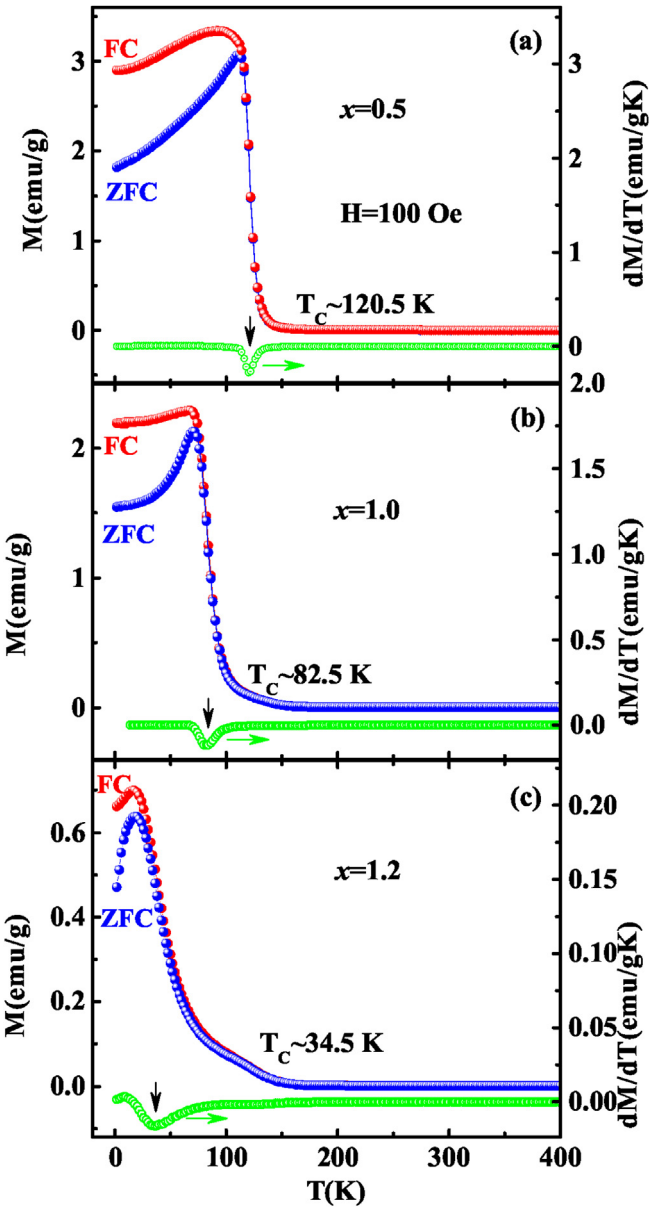


**Fig. 1.** Temperature dependence of magnetization [$M(T)$] under $H = 100$ Oe for $Fe_{1.5-x}Co_xRh_{0.5}Mo_3N$ ($x = 0.5, 1.0, 1.2$).

the Arrott-Noakes equation, $M^{1/\beta}$ vs. $(H/M)^{1/\gamma}$ relations in the high field region should present as a series of straight lines parallel with each other. The relations of $M^{1/\beta}$ vs. $(H/M)^{1/\gamma}$ in the critical temperature region form the modified Arrott plot (MAP).

For a second-order magnetic phase transition, the spontaneous magnetization $M_S$ and initial susceptibility $\chi_0$ can be described by these critical exponents [24,25]:

$$M_S(T) = M_0(-\varepsilon)^{\beta}, \varepsilon < 0, T < T_C \quad (2)$$

$$\chi_0^{-1}(T) = (h_0/M_0)\varepsilon^{\gamma}, \varepsilon > 0, T > T_C \quad (3)$$

where $\varepsilon = (T - T_C)/T_C$ is the reduced temperature; $M_0/h_0$ is critical amplitude. It can be seen that the critical exponent $\beta$ associates with the spontaneous magnetization $M_S$, while $\gamma$ corresponds to the initial susceptibility $\chi_0$.

Due to the characteristic of the three-dimensional crystal (3D)