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Investigation of critical behavior in $Pr_{0.55}Sr_{0.45}MnO_3$ by using the field dependence of magnetic entropy change

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One method of calculating critical exponents based on the field dependence on magnetic entropy change was applied to study critical behavior in $Pr_{0.55}Sr_{0.45}MnO_3$. By using the obtained critical exponents, the modified Arrott plot [A. Arrott and J. E. Noakes, Phys. Rev. Lett. **19**, 786 (1967)] is consistent with that by using conventional method. The calculated critical exponents not only obey the scaling theory, but also inosculate the deduced results from the Kouvel–Fisher method [J. S. Kouvel and M. E. Fisher, Phys. Rev. **136**, A1626 (1964)]. It eliminates the drawback due to utilization of multistep nonlinear fitting in a conventional manner. Therefore, this means can be applied to investigate critical behavior. © *2011 American Institute of Physics*. [doi:10.1063/1.3554390]

The mixed-valent manganites of $RE_{1-x}AE_xMnO_3$ (RE =La to Tb and AE=Ca, Sr, Ba, Pb, etc.) have been extensively investigated on the account of their diverse phase diagrams.[1,2] Earlier works mostly focused on the investigation of phenomena such as colossal magnetoresistance and charge ordering state.[3,4] The double exchange theory and the Jahn–Teller effect were used to understand the ferromagnetic-paramagnetic (FM-PM) phase transition associated with an insulator-metal (IM) transition in the manganites.[5–7] However, more recent investigations have brought forward the ferromagnetic phase transition itself.[8–10] Therefore, for better understanding of this issue, it is necessary to investigate the critical behavior at the phase transition temperature in detail.

The perovskite $Pr_{1-x}Sr_xMnO_3$ is an important member in manganite family with the intermediate one electron bandwidth. In this letter, we select the composition of $Pr_{0.55}Sr_{0.45}MnO_3$, which just lies at the boundary of FM and charge order-antiferromagnetic (CO-AF) phase, to investigate the critical behavior around the PM-FM phase transition temperature.

Generally, the common method to deduce the critical exponents always depends on the fitting values obtained from the Arrott plot according to Arrott–Noakes equation of state, $(H/M)^{1/\gamma}=(T-T_C)/T_C+(M/M_1)^{1/\beta}$,[11] where M_1 is a material constant, and β and γ are the critical exponents. Due to the unknown accurate critical exponents at the beginning of work, the researcher has to apply some different theoretical models to first construct some tentative Arrott plots and then choose the best one for fitting data.[12–14] However, the choice of model and fitting range will directly determine the finial critical values. Different researchers make different choices. Therefore, a considerable uncertainty is unavoidable. To eliminate the drawback, instead, we use the field dependence of magnetic entropy change to deduce the critical exponents. Due to the inner correlation between the critical exponents, they can be calculated from the related equation set. Moreover, in this letter, the calculated exponents not only agree with those deduced from Kouvel–Fisher (KF) method, but also obey the scaling theory,[15,16] indicating that the obtained critical exponents are reliable. Therefore, this method can be widely applied to the similar research field.

Polycrystalline $Pr_{0.55}Sr_{0.45}MnO_3$ was prepared by traditional solid-state reaction method. The structure and phase purity were checked by powder x-ray diffraction (XRD) at room temperature. The XRD patterns showed that the sample was in a single crystallographic phase, without any detectable impurity phase. The temperature dependence of magnetization and the isothermal magnetization were measured by a superconductive quantum interference device.

Figure 1(a) shows the temperature dependence of magnetization (M-T) measured in the magnetic field of 100 Oe. All these data were taken in the warming run after zero-field

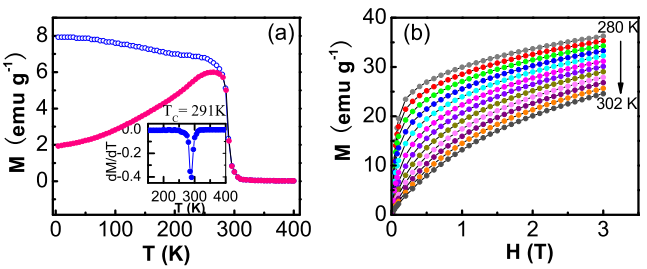

FIG. 1. (Color online) (a) Temperature dependence of magnetization measured at $H=100$ Oe; solid circles are for ZFC, and open circles are for FC; inset shows the plots of dM/dT vs T. (b) Isothermal magnetization vs magnetic field at different temperatures around T_C.

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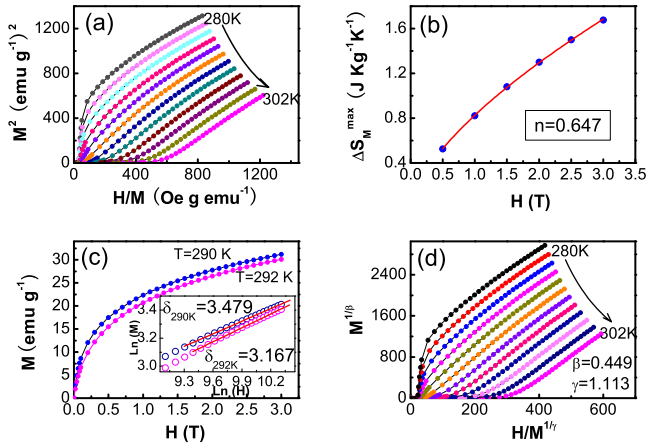

FIG. 2. (Color online) (a) Arrott plot: isotherms of M^2 vs H/M at temperatures around T_C; (b) the maximal magnetic entropy change $|\Delta S_M^{max}|$ vs H (blue circles) and the solid line is the fitting curve; (c) isothermal M vs H plot at 290 and 292 K; the inset shows the same plots in log-log scale, and solid lines are the linear fit following $M_{T_C}=DH^{1/\delta}$; (d) modified Arrott plot: isotherms of $M^{1/\beta}$ vs $(H/M)^{1/\gamma}$ with the calculated $\beta=0.449$ and $\gamma=1.113$.

cooling (ZFC, pink solid circles) and field cooling (FC, blue open circles), respectively. The M-T curve exhibits a sharp PM-FM phase transition. The Curie temperature (T_C), defined by the minimum in dM/dT, has been determined to be $T_C=291$ K [see the inset of Fig. 1(a)]. At low temperatures, the ZFC and FC curves exhibit a considerable divergence, indicating the presence of a spin glass state. Considering the present composition near the boundary of the half-doping composition of $x=0.5$, we need to check whether there exists a CO state. The CO transition is a first-order phase transition and accompanies with an appearance of hysteresis effect in the thermal cycle. Therefore, we measured the M-T curve with the cooling and heating circulation, but no any behavior of thermal hysteresis was observed. Therefore, the sample can be referred to have no CO state, and its PM-FM phase transition should be a second-order phase transition. However, the occurrences of spin glass imply a sign of spin frustration at low temperatures. Therefore, maybe there is a small quantity of short-range CO states in the system which produce some local antiferromagnetic phase to compete with FM phase. Nevertheless, the character of CO state was not detected from the present cooling and heating magnetic measurements probably due to the negligible quantity. Here, in order to further clarify the nature of the FM-PM phase transition, we measure the isothermal magnetization versus applied field around the Curie temperature, which has been shown in Fig. 1(b).

According to the scaling hypothesis,[17] a second-order magnetic phase transition near Curie point is characterized by a set of critical exponents of β, γ, and δ. In order to deduce these parameters, the isothermal magnetization curves (M versus H) should be changed into the so-called Arrott plot, namely, M^2 versus H/M. If the system is in line with the Landau mean-field theory,[17] the relationship of M^2 versus H/M should be shown as a set of parallel straight lines around T_C. Meanwhile, the order of magnetic transition can be determined from the slope of straight line.[18] A positive slope corresponds to the second-order transition, while a negative slope corresponds to a first-order transition. Figure 2(a) is an Arrott plot of M^2 versus H/M. Clearly, in the present case, the positive slope indicates that the phase transition is a second-order PM-FM phase transition, in agreement with the foregoing discussion. However, one can note that all the curves are nonlinear and show downward curvature even at high the field region, indicating that the critical exponents of $\beta=0.5$ and $\gamma=1.0$ are not satisfied. Namely, the mean-field theory cannot be used to describe the critical behavior in the present system. In order to obtain the accurate critical exponents, the conventional method is to use some tentative exponents to construct a new Arrott plot and then fit the data of the linear part or directly fit the data of the initial Arrott plot. After that, the obtained intercepts on the $x(y)$-axis were performed with multistep nonlinear fitting until the finial critical exponents reach steady values. Because of the drawback of this method as mentioned above, we adopted another way to deduce the critical exponents. First, these critical exponents of β, γ, and δ satisfy the Widom scaling relation: $\delta=1+\gamma/\beta$.[19] Meanwhile, the critical exponent of δ is associated with the critical magnetization isotherm at T_C and can be obtained from the following equation: $M_{T_C}=DH^{1/\delta}$,[20] where D is the critical amplitudes. Notably, we need to establish another equation to solve β and γ. In a magnetic system with a second-order phase transition, Oesterreicher and Parker previously proposed a universal relation of the field dependence of magnetic entropy change, namely, $|\Delta S_M^{PK}| \propto H^n$, where $n=2/3$.[21] However, the subsequent experimental results exhibit deviation from $n=2/3$ in the soft magnetic amorphous alloys. Recently, Franco *et al.* further confirmed the existence of the universal relation and reprovided a new relation that agrees better with experimental results:[22]

$$n = 1 + \frac{\beta - 1}{\beta + \gamma}. \tag{1}$$

This relation has been applied to some different universality classes such as the soft magnetic $Fe_{83}Zr_6B_{10}Cu_1$ (Ref. 23) and the cubic Laves phase compounds $TbCo_2$.[24] Thus, if we can know the value of n, the critical exponents of β and γ will be solved. In order to obtain the value of n, we first calculated the magnetic entropy change versus temperature under different magnetic fields using the conventional method .[25] From it, we obtained the relation of $|\Delta S_M^{PK}|$ versus H, as shown by the blue circles in Fig. 2(b). Then, using the above function of $|\Delta S_M^{PK}| \propto H^n$, the value of n was determined to be $n=0.647$. Due to the unknown actual phase transition temperature, we chose two M versus H curves with the nearest temperatures (290 and 292 K) around T_C for calculating δ. As shown in the inset of Fig. 2(c), the values of δ are 3.479 and 3.167 for $T=290$ and 292 K, respectively. Using the obtained values of n and δ, the values of β and γ were calculated to be 0.449 and 1.113 for $T=290$ K, and 0.472 and 1.023 for $T=292$ K, respectively. Thus, a new Arrott plot can be redrawn with the obtained β and γ. Since the calculated critical exponents at $T=292$ K is close to that of mean-field model, there is a delicate difference between the new Arrott plot and Fig. 2(a). Therefore, the critical exponents at $T=292$ K should be excluded. Using the critical exponents $\beta=0.449$ and $\gamma=1.113$ at $T=290$ K, a new Arrott plot has been drawn in the Fig. 2(d). Obviously, a set of parallel lines from 280 to 302 K exhibit in the plot, indicating that the calculated exponents are the popular results.

Nevertheless, the reliability of the calculated β and γ is worth further examining. In the critical region, according to

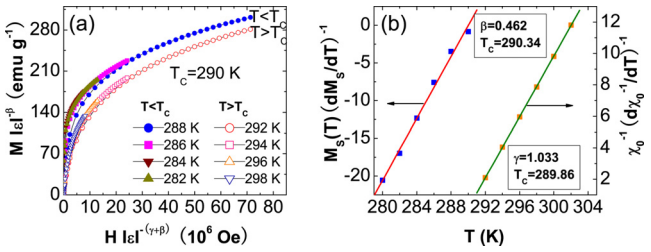

FIG. 3. (Color online) (a) Scaling plots below and above T_C using the calculated exponents of β=0.449 and γ=1.113. (b) Kouvel-Fisher plot for the spontaneous magnetization $M_S(T)$ and the inverse initial susceptibility $\chi_0^{-1}(T)$ [the solid lines are the fitting results with Eqs. (2) and (3)].

the scaling theory, the magnetic equation can be written as $M(H,\varepsilon)\varepsilon^{-\beta}=f_{\pm}(H/\varepsilon^{\beta+\gamma})$, where ε is the reduced temperature $(T-T_C)/T_C$, and f_+ for $T>T_C$ and f_- for $T<T_C$ are regular functions.[16] The equation indicates that $M\varepsilon^{-\beta}$ as a function of $H\varepsilon^{-(\beta+\gamma)}$ yields two universal curves: one for temperature $T>T_C$ and the other for temperature $T<T_C$. As shown in Fig. 3(a), the experimental data fall on two curves, one above T_C and the other below T_C, in agreement with the scaling theory. Therefore, the FM behavior around the Curie temperature was properly renormalized following the scaling equation of state, indicating that the present exponents are reasonably accurate and unambiguous.

In addition, we also used the KF method to deduce the critical exponents for comparison with the critical exponents calculated from field dependence of magnetic entropy change.[15] The KF equations are expressed as follows:

$$\frac{M_s(T)}{dM_s(T)/dT}=\frac{T-T_C}{\beta}, \tag{2}$$

$$\frac{\chi_0^{-1}(T)}{d\chi_0^{-1}(T)/dT}=\frac{T-T_C}{\gamma}. \tag{3}$$

According to Eqs. (2) and (3), $M_s(dM_s/dT)^{-1}$ versus T and $\chi_0^{-1}(d\chi_0^{-1}/dT)^{-1}$ versus T should yield straight lines with slopes $1/\beta$ and $1/\gamma$, respectively. When these straight lines are extrapolated to the ordinate equal to zero, the intercepts on T axis just correspond to T_C. As presented in Fig. 3(b), the fitting results with the KF method give the exponents and T_C to be of β=0.462 with T_C=290.34 K and γ=1.033 with T_C=289.86 K. Obviously, the obtained values of the critical exponents and T_C using the KF method are in agreement with the calculated exponents and the selective T_C=290 K within the experimental precision. Hence, we think that the present calculated method for the study of critical properties is effective and feasible.

In order to further inspect the actual application, we use the above method to calculate the critical exponents of spinel structural selenide $CuCr_2Se_4$. Using the functions of $|\Delta S_M^{PK}| \propto H^n$ and $M_{T_C}=DH^{1/\delta}$, the values of n and δ were determined to be n=0.629 and δ=4.749, respectively. So β and γ are calculated to be β=0.362 and γ=1.349, which are basically in agreement with the values of 0.374 and 1.176 obtained from the conventional fitting method.[26]

At last, we can discuss the reason why the obtained critical exponents have some small deviations from the mean-field theory. Even though $Pr_{0.55}Sr_{0.45}MnO_3$ is a FM material, the phenomenon of disorder affects the critical behavior in this system. The disorder probably arises from dissimilar ions of Pr^{3+} and Sr^{2+} at A site of ABO_3 perovskite structure and the inhomogeneous properties above and below T_C. Therefore, the properties of disorder and inhomogeneity are intrinsic and unavoidable. For $Pr_{0.55}Sr_{0.45}MnO_3$, which just lies at the boundary of FM and CO-AF phase, the impetuous competition between different phases forms a frail balance and causes a large quantum spin fluctuation. In this situation, the system usually reveals an inhomogeneous property which is mainly responsible for the inconsistent critical exponent between the actual material and theoretical model.

In summary, we have studied the critical behavior in $Pr_{0.55}Sr_{0.45}MnO_3$ by using the field dependence of magnetic entropy change. The calculated critical exponents are satisfied with the scaling theory. The simple method effectively avoids the drawback of uncertainty. Therefore, this method can be extensively applied to investigate the critical behavior.

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