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Magnetization Study of the AFM–FM Coexistence in the Manganite System $Pr_{0.5}Ca_{0.5-x}Sr_xMnO_3$

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For the manganites $Pr_{0.5}Ca_{0.5-x}Sr_xMnO_3$ ($x = 0$; 0.3) the temperature and cooling field dependence of the reduced remanence asymmetry indicates the antiferromagnetic exchange between the antiferromagnetic and ferromagnetic spins at the surface of the clusters. For $x = 0.3$ we deduce the radius of the cluster in the field cooled regime to be about 20 times larger than that for $x = 0$.

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1. General information

The manganite perovskites $Pr_{0.5}Ca_{0.5-x}Sr_xMnO_3$ were studied with the aim to investigate magnetic and structural phase transitions as well as the phase separation into the antiferromagnetic (AFM) and ferromagnetic (FM) regions [1, 2]. The neutron diffraction, NMR and magnetometric studies were performed to determine the phase diagram of the system and the relative volumes of the magnetic phases. In the temperature region below about 170 K, these materials represent antiferromagnets containing a small portion ($<$ 10 percents) of the minor ferromagnetic phase. The present contribution follows the works mentioned above and brings new results based on measurements of the exchange bias (EB) [3]. This method, which can yield information on the interface exchange interaction between the phases has been recently applied to a charge ordered manganite $Pr_{1/3}Ca_{2/3}MnO_3$ [4]. We focus our attention on the compositions $x = 0$ and 0.3 representing a small and medium content of the FM phase, respectively.

2. Experimental

The ceramic materials used in the experiments were the same as those described in the work [1]. The samples contained a small volume of Mn_3O_4 hav-

ing $T_N = 44$ K. The measurement of the susceptibility under the applied field $H = 10$ mT and comparison with results for pure Mn_3O_4 enabled us to determine the weight fraction p corresponding to these Mn_3O_4 impurities. This yields $p = 0.0035$ and 0.0016 for the compositions $x = 0$ and $x = 0.3$, respectively. Comparison with the measurements on the $Pr_{0.5}Ca_{0.5}MnO_3$ samples not containing Mn_3O_4 [5] shows that the observed EB shift of the hysteresis loop was not practically affected by the Mn_3O_4 impurity content.

The dc magnetic moment measurements were carried out using a SQUID magnetometer MPMS-5S (Quantum Design). The $m(H)$ loops were measured after cooling the sample with an applied field H_{cool} from room temperature to 5 K and for $T > 5$ K then warmed back to the measuring temperature [4]. In order to eliminate the influence of the training effect we measured 3 subsequent loops and evaluated the results for the last loop. The measured field coold (FC) loops have their centre of symmetry in the plane (H, m) shifted to a point $(-H_E, \delta m)$ with H_E denoting the EB shift and δm a vertical shift of the hysteresis loop. The latter shift has been determined from the maximum and minimum magnetization of the loop using $\delta m = 0.5(m_{max} + m_{min})$. We have verified that the hysteresis loops measured after cooling in the zero field (ZFC) had their centre of symmetry at the point (0,0). The EB shift H_E can be determined from the intersections of the loop with the line $m = \delta m$. The experimental determination of H_E is not so accurate and it is more convenient to characterize the EB effect by the remanence asymmetry defined as $m_E = 0.5[m_{rem}(-) + m_{rem}(+)]$ [4]. The remanences $m_{rem}(-)$ and $m_{rem}(+)$ are given by two intersections of the loop with the m axis. This value was additionally corrected by subtracting the value of δm. The reduced remanence asymmetry $m_{Er} = m_E/m_S$ represents a quantity, which is proportional to the EB shift H_E. In this relation m_S denotes the saturation magnetization depending on temperature and the field H_{cool}.

3. Results and discussion

At $T = 5$ K and $H_{cool} = 2$ T we obtained $m_{Er} = 0.1306$ ($H_E \approx 21.3$ mT) and $x = 0.00567$ ($H_E \approx 1.35$ mT) for $x = 0$ and $x = 0.3$, respectively. The EB shift for the $x = 0$ composition is thus roughly 20 times larger than that for $x = 0.3$. We can imagine that the minor FM phase is formed by spherical clusters with radius R embedded in the AFM matrix. The value of the EB shift should be proportional to surface to volume ratio of these clusters which gives the proportionality $m_{Er} \sim H_E \sim 1/R$. For $H_{cool} = 2$ T thus we may estimate that $R(x = 0.3)/R(x = 0) \approx 20$. The temperature evolution of the reduced remanence asymmetry for $H_{cool} = 2$ T (Fig. 1a) shows a rapid decrease in m_{Er} with increasing temperature so that at $T = 30$ K ($x = 0.3$) and $T = 40$ K ($x = 0.4$) m_{Er} is too small to be measured. The very FM magnetization persists up to the temperatures 170 K, which is near the Néel temperature. At $T \approx 30$–40 K the thermal energy is so large that spins at the surface of the clusters become free and cannot mediate the exchange interaction leading to an EB shift.

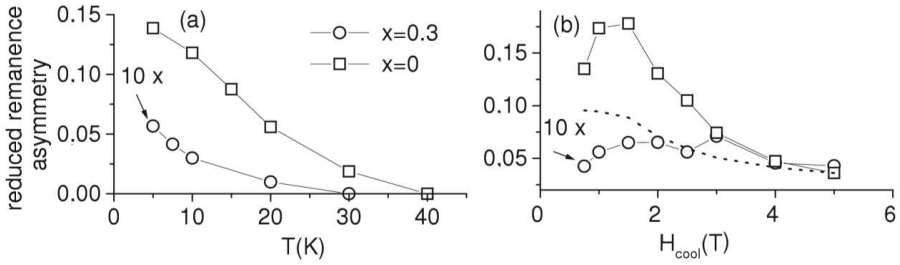

Fig. 1. The reduced remanent asymmetry for $H_{\rm cool} = 2$ T as a function of the temperature (a). The cooling field dependence at $T = 5$ K. For $x = 0$, by the dotted curve, the calculated course proportional to $1/(m_S)^{1/3}$ (b). The values for $x = 0.3$ were multiplied by 10.

The dependence on $H_{\rm cool}$ ($T = 5$ K)(Fig. 1b) can yield information about the sign of the exchange interaction between the FM and AFM spins at the surface of the clusters. The existence of a maximum of m_{Er} and a decrease in m_{Er} with increasing $H_{\rm cool}$ suggests the antiferromagnetic interactions between the spins at the FM–AFM interface [4]. This is valid for a constant volume to surface ratio of the clusters. In our case, we must take into account the variation of this ratio for different values of $H_{\rm cool}$. The volume of the FM phase in the FC regime changes with the cooling field. We assume that the change of the volume is primarily given by a change of the radii of the FM clusters. In this case, the radius of the cluster is proportional to $(m_S)^{1/3}$. For $x = 0$ we plotted the calculated dependence m_{Er} $(H_{\rm cool}) \sim 1/R \sim 1/(m_S)^{1/3}$ with the proportionality constant giving the equality for $H_{\rm cool} = 5$ T (by the dotted curve in Fig. 1b). It can be seen that the experimental dependence m_{Er} $(H_{\rm cool})$ is more expressive than that given by the variation of the radius R. This points to the antiferromagnetic interaction between the FM–AFM spins at the interface.

4. Conclusions

The interface exchange coupling between the AFM and FM spins at the surface of the FM clusters leads to a finite reduced remanence asymmetry. The antiferromagnetic character of this coupling was deduced taking into account the probable dependence of the cluster radius on the cooling field. For $H = 2$ T the cluster radius of the manganite containing Sr is estimated to be 20 times larger than that of $Pr_{0.5}Ca_{0.5}MnO_3$.

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