$\begin{array}{l} \text{MAGNETIC PROPERTIES OF } \Pr_{1-x} \mathrm{Sr}_{1+x} \mathrm{MnO}_4 \\ (0.3 \leq x \leq 0.8) \text{ SINGLE CRYSTALS}^{* \ **} \end{array}$

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We report the magnetic properties of $Pr_{1-x}Sr_{1+x}MnO_4$ single crystals in the composition range $0.3 \leq x \leq 0.8$. All the compositions studied show highly anisotropic spin-glass type magnetic ordering at low temperatures. Some of the compositions (x > 0.6) exhibit a broad maximum in the magnetic susceptibility above 100 K, highlighting the 2D magnetic interactions in these compounds. A possible charge ordering transition is observed around 265 K for the x = 0.8 compound.

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1. Introduction

The hole-doped single layer compounds $\text{La}_{1-x}\text{Sr}_{1+x}\text{MnO}_4$, that crystallizes in the K₂NiF₄ structure, have been investigated by several groups [1–3]. The x = 0.5 compound exhibits charge/orbital ordering at ~230 K and a variety of magnetic ordering at lower temperatures. We present the preparation of single crystals of the isostructural compound, $\text{Pr}_{1-x}\text{Sr}_{1+x}\text{MnO}_4$ $(0.3 \leq x \leq 0.8)$ and the detailed investigation of their magnetic properties.

2. Experimental

 $Pr_{1-x}Sr_{1+x}MnO_4$ single crystals were prepared by the floating zone method using a four-mirror infrared image furnace. The crystal growth was performed under different gas atmospheres of oxygen/argon for different

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compositions [4]. The magnetization was measured using a SQUID Magnetometer (MPMS, Quantum Design, USA). Both the sample geometries, H|| ab and H|| c, were used for the measurements.

3. Results and discussions

The well studied compound, $La_{0.5}Sr_{1.5}MnO_4$ has been shown to exhibit simultaneous charge [1,2] and orbital ordering [3] above the magnetic ordering temperature. The charge ordering is seen as an explicit discontinuity in the susceptibility at around 230 K. However, the magnetic susceptibility of $Pr_{0.5}Sr_{1.5}MnO_4$ (x = 0.5) does not reveal any anomaly related to charge ordering (see Fig. 1). The magnetic susceptibility goes through a broad maximum around 130 K, and shows a magnetic ordering at low temperatures (~20 K). The magnetic ordering temperature is found to be dependent



Fig. 1. ZFC and FC magnetic susceptibility as a function of temperature for $Pr_{0.5}Sr_{1.5}MnO_4$ (x = 0.5) compound for H||ab and H||c (inset) at two different applied fields, H = 1 kOe and 10 kOe.

on the applied magnetic field, but the broad hump in susceptibility is independent of the magnetic field. The broad maximum in susceptibility is also shown by other compositions with x = 0.6 and 0.7. These broad maxima in susceptibility may correspond to strong 2D magnetic interactions in these compounds. The magnetic susceptibility of $Pr_{0.2}Sr_{1.8}MnO_4$ (x = 0.8) as a function of temperature is shown in Fig. 2. The susceptibility shows an anomaly around 265 K, which is reminiscent of a charge ordering type behavior. This anomaly is present in both ZFC and FC data in both the sample geometries. The anomaly at lower temperature (~50 K) may correspond to a spin-glass type magnetic ordering generally observed in these type of compounds. Preliminary specific heat measurement analysis shows that the anomaly in the susceptibility at 265 K manifests itself as a change in the slope of the heat capacity vs temperature curve [4]. The charge ordering behavior observed in $Pr_{0.2}Sr_{1.8}MnO_4$ is similar to that found in $Pr_{0.25}Ca_{1.75}MnO_4$ [5], where the high resolution electron diffraction measurements reveal a "1-3" Mn^{3+}/Mn^{4+} charge ordering. Detailed electron diffraction and neutron diffraction measurements are required to ascertain the exact nature of the charge ordering in $Pr_{0.2}Sr_{1.8}MnO_4$.



Fig. 2. Temperature variation of the ZFC and FC magnetic susceptibility for $Pr_{0.2}Sr_{1.8}MnO_4(x = 0.8)$ for H||ab and H||c (inset).

All the $\Pr_{1-x} \operatorname{Sr}_{1+x} \operatorname{MnO}_4$ compounds $(0.3 \leq x \leq 0.8)$ show spin-glass type magnetic ordering at low temperatures that can be determined from the difference between the ZFC (zero field cooled) and FC (field cooled) susceptibility below the magnetic ordering from Fig. 1 as well as in Fig. 2. Further evidence for the development of a small magnetic moment at low temperatures is evident from the field dependence of magnetization. Fig. 3 shows a typical M vs H curve for the x = 0.3 compound. At 2 K (for H||c), a clear hysteresis exists with a non-zero remnant magnetization. However, no such hysteresis exists for H||ab. It is interesting to note that the magnetic properties are highly anisotropic, highlighting the 2D nature of these compounds. The nature of magnetic ordering and the susceptibility values are very much different in the two orientations, as evident from Figs 1–2.



Fig. 3. Magnetization as a function of applied field for $Pr_{0.7}Sr_{1.3}MnO_4$ (x = 0.3) for H||c and H||ab (inset).

In conclusion, we have shown that $Pr_{1-x}Sr_{1+x}MnO_4$ compounds show anisotropic magnetic properties in accordance with their 2D nature. They exhibit spin-glass type magnetic ordering at low temperatures. A possible charge ordering is seen at 265 K for the x = 0.8 compound.

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