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Journal of Magnetism and Magnetic Materials 196–197 (1999) 487–489

Journal of
Magnetism
and
Magnetic
materials

Spin dynamics of $\text{La}_{0.85}\text{Sr}_{0.15}\text{CoO}_3$ perovskite

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Abstract

Dynamic magnetic properties of $\text{La}_{0.85}\text{Sr}_{0.15}\text{CoO}_3$ have been experimentally studied by AC-susceptibility and time-dependent zero-field-cooled magnetisation experiments. The onset of ferromagnetic order appears at $T_C \approx 240$ K and a frequency-dependent maximum of both components of the AC-susceptibility is observed at $T \approx 60$ K indicating a low-temperature re-entrant spin glass phase. However, a dynamic scaling analysis of the behaviour does not yield critical slowing down but thermally activated dynamics and thus no finite temperature transition to a re-entrant spin-glass phase. Magnetic ageing, a signature of frustrated and disordered magnetic systems, is found to exist both above and below the frequency dependent maximum in the susceptibility. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Perovskites; Re-entrant ferromagnets; Spin glasses; Ageing; Magnetic susceptibility; $\text{La}_{0.85}\text{Sr}_{0.15}\text{CoO}_3$

The interest in magnetic and transport properties of $\text{Ln}_{1-x}\text{B}_x\text{MO}_3$ (Ln = lanthanide, B = alkaline earth, M = 3d transition metal) perovskites has increased immensely after the observation of colossal magnetoresistance (CMR) in samples with M = Mn [1,2]. Co-based $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$ perovskites do not show a very high magnetoresistance [3,4], but they still show interesting magnetic properties [5] caused by a change of the spin state of the Co^{3+} ions with temperature. An abrupt decrease of the susceptibility below 100 K in LaCoO_3 is explained by a small energy difference between the low-spin (t^6e^0 ; $S = 0$) and high-spin (t^4e^2 ; $S = 2$) configurations (of the order of ≈ 0.03 eV) which allows a gradual thermal population of the high spin state with increasing temperature up to about 100 K, where an equilibrium 50:50 population of the two states is reached [6,7]. Sr doping leads to a phase segregation into hole-rich ferromagnetic metallic clusters and a

hole-poor LaCoO_3 -like matrix. This matrix may mediate a temperature-dependent intercluster interaction. At lower temperatures the LaCoO_3 -like matrix would tend to the $S = 0$ configuration, weakening the interaction in between the clusters. This tendency leads to an increased disorder which may cause a re-entrant spin glass phase. In a previous work on $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$ [8], two anomalies in the temperature dependence of the AC-susceptibility were observed and ascribed to the blocking temperature of two different interaction regimes. In this paper, we report AC-susceptibility and time dependent magnetisation measurements on $\text{La}_{0.85}\text{Sr}_{0.15}\text{CoO}_3$ which reveal an ageing behaviour and distinguishes that a true re-entrant spin-glass transition does not occur.

Sample synthesis and characterisation are described elsewhere [5]. The AC-susceptibility was measured in a LakeShore AC-susceptometer at different frequencies and in an AC-field of 5 Oe. In Fig. 1a and Fig. 1b the real (χ') and imaginary (χ'') parts of the AC-susceptibility are plotted versus temperature for three different frequencies. A frequency dependence of the response is seen at all temperatures below T_C , both in the in-phase and the out-of-phase components. The onset of ferromagnetic

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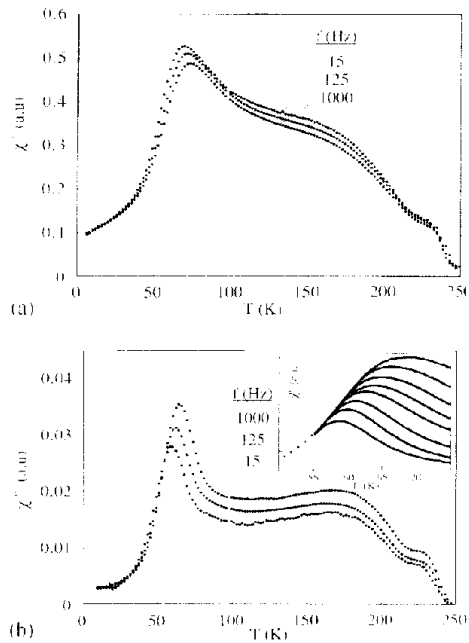


Fig. 1. (a) Real (χ') and (b) imaginary (χ'') parts of the AC susceptibility for $\omega/2\pi = 15$, 125 and 1000 Hz and AC-field $h = 5$ Oe. The inset displays χ'' for $\omega/2\pi = 0.51$, 1.7, 5.1, 17, 51, 170, 510, 1700 Hz [bottom to top], AC-field $h = 0.5$ mOe.

order occurs at $T_C \approx 240$ K and is seen as a sharp increase of both components of the susceptibility. This is in agreement with neutron diffraction results, which evidence the set up of ferromagnetic order at a similar temperature [9]. A spinodal phase segregation into Sr-rich and Sr-poor regions due to low-temperature synthesis might be the origin of such an effect. The ferromagnetic transition is followed by a shoulder (weak maximum in the out-of-phase component) at about 170 K and a strongly frequency-dependent main maximum in both components at $T \approx 60$ K that is shifted towards lower temperatures with decreasing frequency. This behaviour of the AC susceptibility somewhat resembles that of a conventional re-entrant ferromagnet [10]. To carefully investigate the frequency dependence of the main maximum a non-commercial SQUID magnetometer was used. In the inset of Fig. 1b, the out-of-phase component of the susceptibility, measured in an AC-field of 0.5 mOe, is shown at different frequencies in the range $0.51 \leq f(\text{Hz}) \leq 1700$. The position of the maximum in the out-of-phase component can be used to define the freezing temperature $T_f(\omega)$ and the characteristic relaxation time of the system at this maximum corresponds to the observation time $t \approx 1/\omega$ of the AC-field. An analysis of the behaviour of $T_f(\omega)$ shows that conventional critical slowing down is not applicable but that the slowing down of the dynamics follows a modified Arrhenius law, $\log(t/t_0) \propto T_f^{-2}$ with $t_0 = 10^{-14}$ s. Our results thus indi-

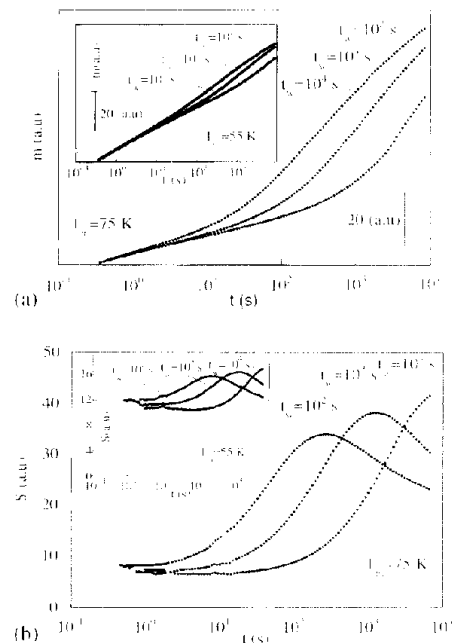


Fig. 2. ZFC magnetisation, $m(t)$ versus $\log t$ (a), and relaxation rate $S(t) = (1/h) dM/d \log t$ versus $\log t$ (b), for $t_w = 100$, 1000, 10 000 s, at $T_m = 75$ K and $h = 1$ Oe. The insets show the corresponding curves at $T_m = 55$ K.

cate that there is no finite temperature spin-glass transition T_g in $\text{La}_{0.85}\text{Sr}_{0.15}\text{CoO}_3$ in some contrast to a previously reported result [11].

To reveal if the system exhibits ageing, we performed time-dependent zero-field-cooled (ZFC) relaxation measurements. The system is then cooled down with a cooling rate of 3 K/min from a reference temperature, here $T_{\text{ref}} = 280$ K, to a measurement temperature, T_m , where the sample is kept at a wait time of t_w . Thereafter a weak DC-field, h , is applied and the magnetisation, m , is recorded as a function of time. In Fig. 2a m is plotted versus $\log t$ for $T_m = 75$ K and $T_m = 55$ K (in the inset) for $t_w = 100$, 1000 and 10 000 s. The corresponding relaxation rate, $S(t) = (1/h) dM/d \log t$, is plotted in Fig. 2b. A strong wait time dependence of the response is observed at both temperatures. The m versus $\log t$ curves have an inflection point when the observation time nearly equals t_w and the relaxation rate attains a corresponding maximum, thus a distinct ageing behaviour is observed. Comparing the relaxation rates at $T_m = 55$ K and $T_m = 75$ K a quantitative difference is observed. The influence of ageing on the relaxation rate is stronger above the $T_f(\omega)$ than below, e.g. the ratio between the maximum at $t \approx t_w$ and the level at $t \approx 1$ s, for the curves measured with $t_w = 1000$ s, is about 5.5 at temperatures above and 1.5 at temperature below $T_f(\omega)$. A similar behaviour has also been observed in a conventional re-entrant ferromagnet [12].

The experiments show that the compound $\text{La}_{0.85}\text{Sr}_{0.15}\text{CoO}_3$ exhibits ageing, a signature of disorder and frustration, both above and below $T_f(\omega)$. The system shows some striking similarities with a conventional re-entrant ferromagnet, but the slowing down of the dynamics does not support the existence of a (re-entrant) spin-glass phase at low temperatures.

Financial support from the Swedish Natural Science Research Council (NFR) and Spanish DGICYT MAT98-0416 is acknowledged.

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