AIP | Journal of Applied Physics

Magnetic frustration effects in LaCaMnO 3 single crystals

Manh-Huong Phan, V. Srinivas, Seong-Cho Yu, and Nam Hwi Hur

Citation: Journal of Applied Physics **93**, 8200 (2003); doi: 10.1063/1.1558589
View online: http://dx.doi.org/10.1063/1.1558589
View Table of Contents: http://scitation.aip.org/content/aip/journal/jap/93/10?ver=pdfcov
Published by the AIP Publishing

Magnetic frustration effects in LaCaMnO$_3$ single crystals

Manh-Huong Phan
Department of Physics, Chungbuk National University, Cheongju, 361-763, Korea

V. Srinivas
Department of Physics, Chungbuk National University, Cheongju 361-763, Korea and Department of Physics and Meteorology, Indian Institute of Technology, Kharagpur-721 302, West Bengal, India

Seong-Cho Yu[a)]
Department of Physics, Chungbuk National University, Cheongju, 361-763, Korea

Nam Hwi Hur
Center for CMR Materials, Korea Research Institute of Standards and Science, Yusong, P.O. Box 102, Taejon 305-600, Korea

(Presented on 14 November 2002)

We investigated dc and ac susceptibilities of $La_{1-x}Ca_xMnO_3$ ($x=0.2,0.3$) single crystals. Obtained results show that $La_{0.8}Ca_{0.2}MnO_3$ is clearly magnetically more frustrated than $La_{0.7}Ca_{0.3}MnO_3$, indicating that charge/orbital fluctuation is enhanced in $La_{0.8}Ca_{0.2}MnO_3$. A remarkable magnetic anomaly in the imaginary component of the ac susceptibility appears around 100 K for $La_{0.8}Ca_{0.2}MnO_3$ to be attributed to the magnetic frustration arising from the strong competition between antiferromagnetic and ferromagnetic interactions that favor the existence of short-range charge/orbital ordering in this sample. In connection with the resistivity data, we propose that the order parameter of magnetic origin should be coupled to the other degrees of freedom in the case of $La_{0.8}Ca_{0.2}MnO_3$ rather than $La_{0.7}Ca_{0.3}MnO_3$.
[DOI: 10.1063/1.1558589]

Recently, there has been much interest in the close interplay between magnetism and transport properties in $La_{1-x}Ca_xMnO_3$ compounds.[1–4] As reported recently in $La_{1-x}Ca_xMnO_3$ ($x=0.2,0.3$) single crystals,[3,4] we have found that $La_{0.8}Ca_{0.2}MnO_3$ becomes an insulator and has a smooth rise in resistivity with decreasing temperature below T_C. Unlike the transport behavior of $La_{0.8}Ca_{0.2}MnO_3$, below the metal-insulator transition temperature (T_{MI}) $La_{0.7}Ca_{0.3}MnO_3$ exhibits metallic character judging from positive values in $d\rho/dT$. On the magnetic characteristics, it is remarkable that there is a prominent deviation of the field-cooled (FC) magnetization from the zero-field-cooled (ZFC) magnetization below T_C in both compositions, which have already been reported in Ref. 4 and are re-shown here in Fig. 1. Moreover, to our surprise, a magnetic anomaly occurred around 100 K in the ZFC magnetization (M_{ZFC}) curve of $La_{0.8}Ca_{0.2}MnO_3$ involving probably charge-ordered phase transition was observed. The large deviation of the FC magnetization from the ZFC magnetization of both compositions and the magnetic anomaly in M_{ZFC} curve of $La_{0.8}Ca_{0.2}MnO_3$ could be attributed to magnetic frustration arising from the competition between antiferromagnetic (AFM) and ferromagnetic (FM) interactions. However, this argument had been left as an open question. Thus it is desirable to further clarify the magnetic frustration effects in the $La_{1-x}Ca_xMnO_3$ ($x=0.2,0.3$) single crystals.

In the present work, we tried to scrutinize the magnetic frustration effects in the $La_{1-x}Ca_xMnO_3$ ($x=0.2,0.3$) single crystals from dc and ac susceptibility measurements.

[a)]Electronic mail: scyu@chungbuk.ac.kr

$La_{1-x}Ca_xMnO_3$ ($x=0.2,0.3$) single crystals were prepared by the floating zone method using an infrared radiation convergence-type image furnace that consists of four mirrors and halogen lamps; details of the growth conditions have been described elsewhere.[5] The starting ceramic rods were obtained from the solid-state reaction of La_2O_3, $CaCO_3$, and $MnCO_3$ with a stoichiometric ratio. X-ray diffraction data and electron-probe microanalysis confirmed the quality of the crystals. The magnetic measurements were performed using a Quantum Design MPMS-5 superconducting quantum interference device magnetometer or a PPMS-7 magnetometer. The measurements of real and imaginary parts of the ac susceptibilities for the alloy were carried out from 70 to 300 K by using the ACS-7000 susceptometer. The resistivity measurements were carried out with a standard four-probe method in zero magnetic fields. Electrical contacts were made with silver paint.

In order to clarify the magnetic frustration effects in $La_{1-x}Ca_xMnO_3$ ($x=0.2,0.3$) single crystals, the dc and ac susceptibility measurements for both samples have been carried out. The real (χ') and imaginary (χ'') components of the ac susceptibility of $La_{0.7}Ca_{0.3}MnO_3$ sample are shown in Fig. 2. As one can see from Fig. 2, $La_{0.7}Ca_{0.3}MnO_3$ obviously exhibits a sharp phase transition at T_C. This transition prefers a first-order insulator-paramagnetic to metal-ferromagnetic transition, which is consistent with the magnetization data that have been reported earlier in Refs. 4 and 6. It is important to note that a slight decrease in χ' and χ'' for $La_{0.7}Ca_{0.3}MnO_3$ at temperatures below T_C means that this sample belongs to a ferromagnetic metal.[3,4,6] Similar behaviors in the resistivity and χ'' are also found on the single

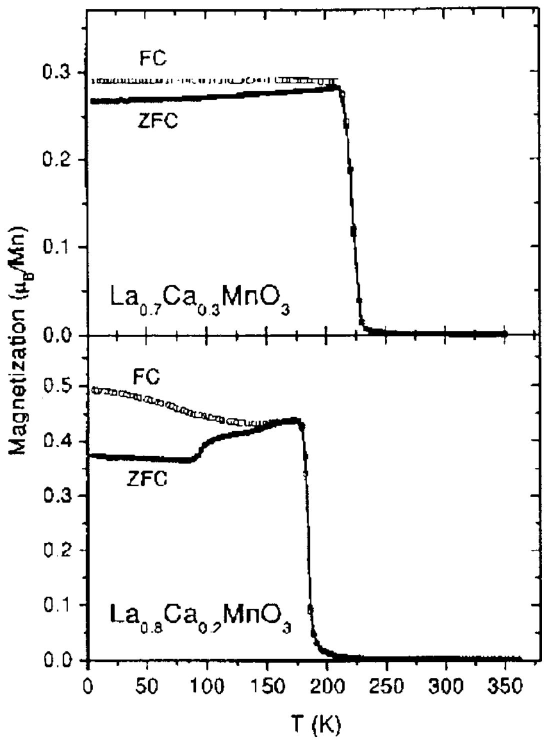

FIG. 1. Top and bottom panels show temperature dependence of magnetization of $La_{0.7}Ca_{0.3}MnO_3$ and $La_{0.8}Ca_{0.2}MnO_3$, respectively.

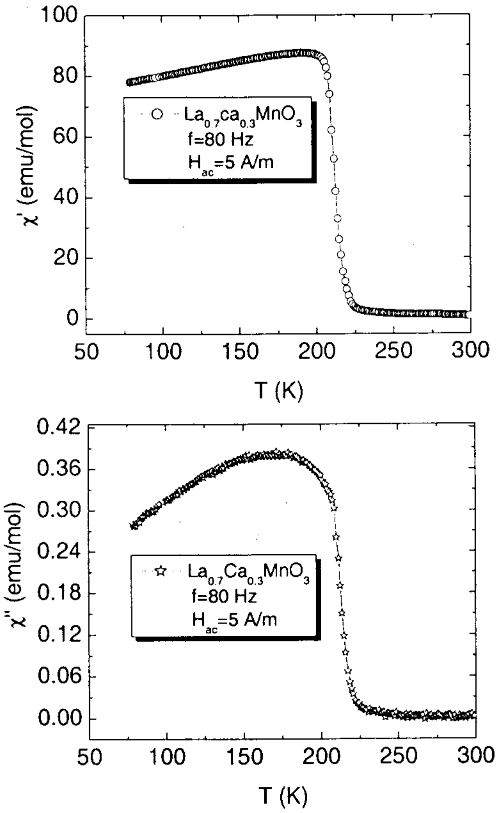

FIG. 2. The real (χ', top panel) and imaginary (χ'', bottom panel) components of ac susceptibility for the $La_{0.7}Ca_{0.3}MnO_3$ sample measured at 80 Hz using 5 A/m ac field with zero biasing dc field.

crystal of $Nd_{0.7}Sr_{0.3}MnO_3$.[7] For $Nd_{0.7}Sr_{0.3}MnO_3$, however, a broad hump in χ'', starting to develop at T_C, appeared and even more remarkably the hump is still present down to the lowest measured temperature. The difference in χ'' at temperatures below T_C between the $La_{0.7}Ca_{0.3}MnO_3$ and $Nd_{0.7}Sr_{0.3}MnO_3$ single crystals can be attributed to different changes in the Mn–O–Mn bond angle and the Mn–O bond distance arising from the A-site average ion radius being bigger for La than for Nd. For comparison, we measured $\chi'(T)$ and $\chi''(T)$ curves of $La_{0.8}Ca_{0.2}MnO_3$ as well as $La_{0.7}Ca_{0.3}MnO_3$. As displayed in the top panel of Fig. 3, a sharp peak in $\chi'(T)$ of $La_{0.8}Ca_{0.2}MnO_3$, measured at $f = 80$ Hz and $H_{ac} = 5$ A/m, appeared at $T \sim 170$ K. This temperature is very close to the T_C and the T_{MI}.[3,4] Despite the time-dependent dynamics due to the blocking process of the clusters, we should recall that the temperature dependence of $\chi'(T)$ including the maximum at $T \sim 170$ K might be qualitatively explained similarly as we have dealt with $M_{ZFC}(T)$ implying a resemblance of this maximum to the cusp observed in the ZFC magnetization curves for $La_{0.8}Ca_{0.2}MnO_3$.[4] We observed that this peak of $\chi'(T)$ is shifted with increasing frequency, indicating a spin-glass-like behavior.[7,8] On the basis of the $\chi'(T)$ curves observed in Fig. 3, it is confirmed that $La_{0.8}Ca_{0.2}MnO_3$ has a spin-glass-like feature in an insulating state at low temperatures below 150 K, and in a metallic one at higher temperatures (up to 176 K). Unlike $La_{0.8}Ca_{0.2}MnO_3$, the single crystals of $La_{0.7}Ca_{0.3}MnO_3$, $Nd_{0.7}Sr_{0.3}MnO_3$, and $Pr_{0.63}Sr_{0.37}MnO_3$ all display a frequency-dependent behavior in the metallic phase at temperatures below T_C.[7] $La_{0.8}Ca_{0.2}MnO_3$ favors a short-range charge/orbital ordering, which arises from the strong competition between AFM and FM interactions. This is connected with the resistivity data of the sample, in which the short-range ordering is likely to respond for the upturn of resistivity below T_C.[3] Furthermore, we measured χ' in the dc external magnetic fields of 5, 10, and 20 A/m and found that χ' is independent of the dc external magnetic field. It reflects that a strong electron-lattice coupling that occurs in the low temperature region below T_C was not affected by the application of dc magnetic fields. For a thorough understanding of the spin dynamics of $La_{0.8}Ca_{0.2}MnO_3$, we measured the imaginary component of ac susceptibility (χ'') at the same frequencies as for the real component. In the top panel of Fig. 4, χ'' for $La_{0.8}Ca_{0.2}MnO_3$ (measured at $f = 80$ Hz) is presented. Surprisingly, χ'' appeared to have two peaks between 70 and 176 K. One peak appeared at $T \sim 170$ K and that coincides with the peak observed from χ', the other peak is found at $T \sim 83$ K. On the other hand, it has been shown that $\chi''(T)$ reflects the magnetic energy dissipation in the sample and is proportional to the area of the hysteresis loop within one period of the ac field at an equilibrium temperature.[7] Therefore, from the different behavior in χ'' at temperatures below T_C between $La_{0.7}Ca_{0.3}MnO_3$ (Fig. 2) and $La_{0.8}Ca_{0.2}MnO_3$ (Fig. 4), we confirm that the spin dynamics of the two samples are very different. In other words, energy dissipation of $La_{0.8}Ca_{0.2}MnO_3$ is much stronger than that of $La_{0.7}Ca_{0.3}MnO_3$.[7,9] Here, it should be noted that both χ' and χ'' for $La_{0.8}Ca_{0.2}MnO_3$ and $La_{0.7}Ca_{0.3}MnO_3$ did not completely vanish below T_C, while $LaMnO_3$ has χ'' that disappears almost completely below T_N.[7] It implies that the mag-

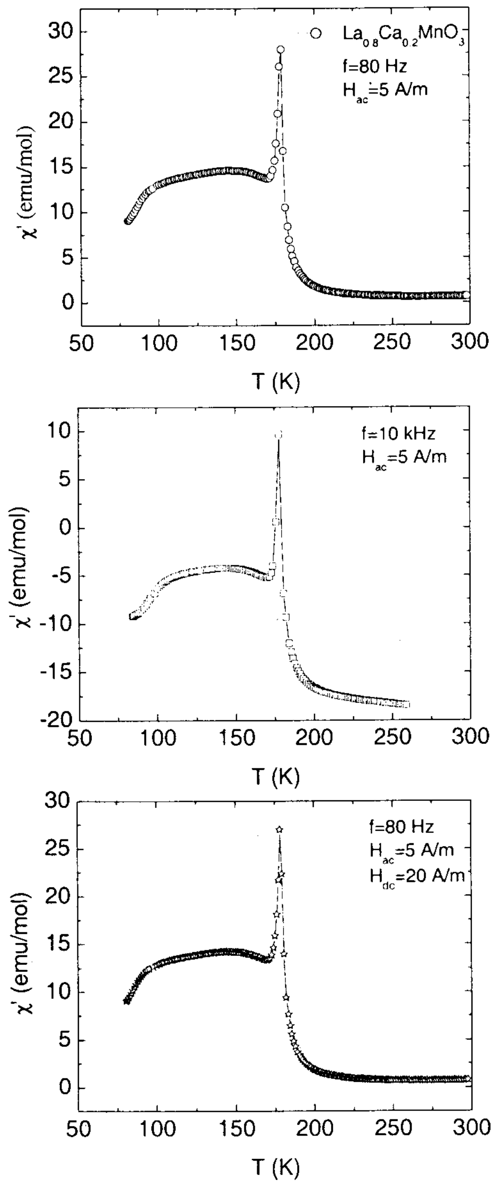

FIG. 3. The real (χ') component of ac susceptibility for the $La_{0.8}Ca_{0.2}MnO_3$ sample measured at $f=80$ Hz, $H_{ac}=5$ A/m, $H_{dc}=0$ A/m (top panel); at $f=10$ kHz, $H_{ac}=5$ A/m, $H_{dc}=0$ A/m (middle panel); at $f=80$ Hz, $H_{ac}=5$ A/m, $H_{dc}=20$ A/m (bottom panel).

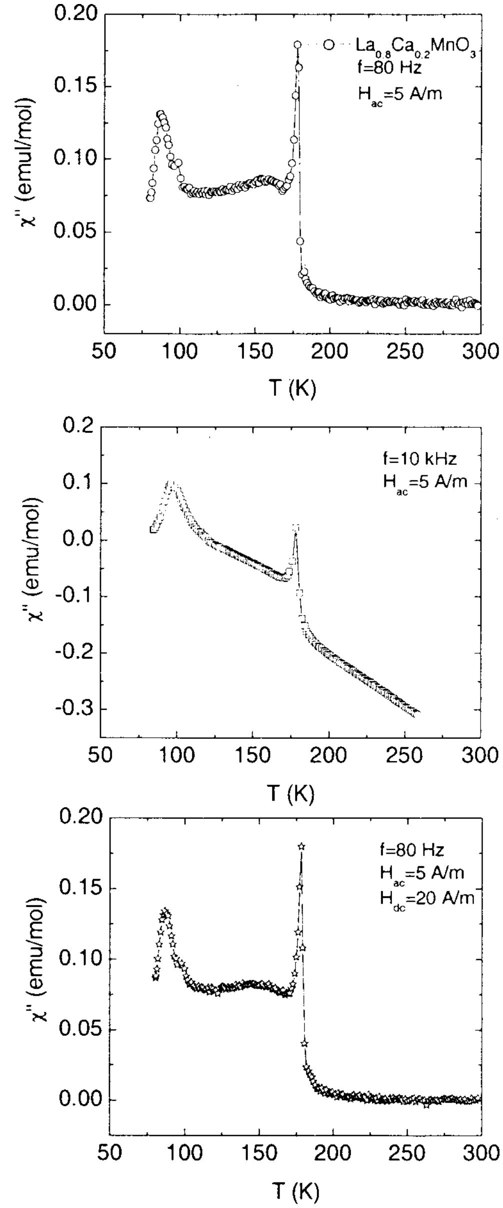

FIG. 4. The imaginary (χ'') component of ac susceptibility for the $La_{0.8}Ca_{0.2}MnO_3$ sample measured at $f=80$ Hz, $H_{ac}=5$ A/m, $H_{dc}=0$ A/m (top panel); at $f=10$ kHz, $H_{ac}=5$ A/m, $H_{dc}=0$ A/m (middle panel); at $f=80$ Hz, $H_{ac}=5$ A/m, $H_{dc}=20$ A/m (bottom panel).

netic frustration effects are mainly considered in strongly electron-correlation systems, and depend on the doping concentration in the sample. Hence, in the case of $La_{0.8}Ca_{0.2}MnO_3$ a magnetic anomaly in χ'' around 100 K is reported. We should recall that the magnetic anomaly in the ZFC magnetization curve of $La_{0.8}Ca_{0.2}MnO_3$ occurred around 100 K, as discussed earlier in Ref. 4 and re-shown in Fig. 1. As one can see from Fig. 4, at $f=80$ Hz, the anomaly in χ'' remains unchanged as $H_{dc}=20$ A/m is applied, while it disappeared at a higher frequency of 10 kHz. Thereby, we conclude that the magnetic transition around 100 K involving probably charge-ordered phase is affected by the frequency. We note that, at $f=10$ kHz, both peaks observed in $\chi''(T)$ for $La_{0.8}Ca_{0.2}MnO_3$ at $T\sim 170$ K and ~ 83 K are strongly modified. It means that the spin dynamics of $La_{0.8}Ca_{0.2}MnO_3$ are drastically changed at high frequencies (~ 10 kHz).

The authors would like to thank Professor Sunk Kun Oh for helpful discussions. Research at the Chungbuk National University was supported by Korea Research Foundation Grant No. KRF-2001-005-D20010.

[1] D. L. Huber *et al.*, Phys. Rev. B **60**, 12155 (1999).
[2] V. A. Atsarkin *et al.*, Phys. Rev. B **63**, 092405 (2001).
[3] H. Yi, C. S. Hong, and N. H. Hur, Solid State Commun. **114**, 579 (2000).
[4] C. S. Hong, W. S. Kim, and N. H. Hur, Phys. Rev. B **63**, 092504 (2001).
[5] C. S. Hong *et al.*, Chem. Mater. (to be published).
[6] H. Y. Hwang *et al.*, Phys. Rev. Lett. **75**, 914 (1995).
[7] J. G. Park *et al.*, Phys. Rev. B **60**, 14804 (1999).
[8] J. M. De Teresa*et al.*, Phys. Rev. Lett. **76**, 3392 (1996).
[9] D. N. H. Nam *et al.*, Phys. Rev. B **59**, 4189 (1999).