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2008 Europhys. Lett. 82 37005

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Microwave detection of magnetic phase avalanches in La$_{0.225}$Pr$_{0.4}$Ca$_{0.375}$MnO$_3$ manganites

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received 9 January 2008; accepted in final form 13 March 2008
published online 22 April 2008

PACS 75.10.-b – General theory and models of magnetic ordering
PACS 78.70.Gq – Microwave and radio-frequency interactions
PACS 45.70.Ht – Avalanches

Abstract – Recent low-temperature studies on La$_{5/8-y}$Pr$_y$Ca$_{3/8}$MnO$_3$ ($y = 3/8$) have shown that the occurrence of the first-order transition in these family of perovskites manifests itself in the form of magnetic and electric avalanches. In this paper we report on the detection of such avalanches performing microwave experiments with coaxial resonators (low frequency $\sim 3$ GHz) and cavities (high frequency $\sim 50$ GHz) at different temperatures and magnetic fields. Short measuring times enabled us to detect the rapid variation in the energy absorption and/or cavity perturbation of the microwaves due to the occurrence of these phase transitions.

Introduction. – Recently, the origin of the colossal magnetoresistance (CMR) in perovskite manganites has been the subject of intensive research work [1,2]. Manganites as La$_{5/8-y}$Pr$_y$Ca$_{3/8}$MnO$_3$ show a wide range of fascinating properties connecting the spins with the electrical charges and their spatial and geometrical arrangements. One of the most interesting points is the coexistence of phase separated (PS) states: charge disordered ferromagnetic (CD-FM) and charge ordered antiferromagnetic (CO-AFM) [3,4]. In fact, a surprising property that has been recently found in manganites is the abrupt and rapid phase change of the entire sample from the CO-AFM phase to the CD-FM that takes place at temperatures below 6 K under the effect of a fast sweeping magnetic field, corresponding to a front propagating through the sample [5]. These avalanche processes involve heat propagation, as those reported on molecule magnets [6,7], and consequently they only occur when the thermal coupling with the environment is not very good or the sample is large enough to ensure the spins do not have time to thermalise. Since the CO-AFM phase is insulating and the CD-FM phase is conducting, the phase transition avalanches are accompanied by the occurrence of colossal resistance avalanches. It is reported that both magnetic and electric avalanches in LaPrCaMnO samples are associated to some kind of percolation process [8]. Indeed, most of the resistivity measurements performed until now involved either MFM images or four-wire probes, and consequently, they are strongly affected by the local configuration and creation of single paths, thus not reflecting at all the bulk behaviour [9,10]. Microwave studies on manganites are, however, scarcely found, and mainly concerned with the high-temperature (order-disorder) transitions [11–14]. In fact, the use of microwave techniques in the study of phase transitions in manganites opens a new perspective on this phenomenon. This letter describes experiments in which the phase transitions in La$_{0.225}$Pr$_{0.4}$Ca$_{0.375}$MnO$_3$ are detected via microwave techniques in order to better understand the bulk behaviour when the CMR avalanche takes place. Two kinds of experiments are reported; in the first set, we used microwave frequencies in the S-band ($\sim 3$ GHz) and a coaxial resonator whereas in the second set, high-frequency band (50–60 GHz) experiments were performed using electromagnetic cavities.

Low frequency. – In the first experimental setup a sample of La$_{0.225}$Pr$_{0.4}$Ca$_{0.375}$MnO$_3$ with dimensions $1 \text{mm} \times 1.2 \text{mm} \times 1.6 \text{mm}$, was placed inside a coaxial resonator short-circuited at both ends, avoiding contact with the metallic parts. The system was 31.5 mm long and 4 mm wide. The resonator was placed in a Quantum Design commercial magnetometer system working at temperatures in the range of 2 K to 300 K, capable of
producing fields up to 5 T. RF detection measurements at the fundamental frequency $f = 3.85$ GHz were performed using an Agilent portable network analyser (PNA), and correspond to the signal reflected ($S_{11}$) from the coaxial resonator.

**Results.** $\text{La}_{0.225}\text{Pr}_{0.4}\text{Ca}_{0.375}\text{MnO}_3$ shows two metastable states, characterized by different proportions of magnetic phases depending on the percolation process that the ferromagnetic domains have followed. After zero-field cooling (ZFC) the system is blocked in a metastable state with a predominance of the CO-AFM insulating phase. The increase in temperature promotes the dynamics of the growing of the CD-FM domains over the antiferromagnet until $T_B = 25$ K, where the PS becomes completely dynamic and the CD-FM domains would finally be the majority of the sample. A transition temperature $T_C$ from the CD-FM conducting phase to the insulating CO-AFM is seen at higher temperatures, between 60 K and 100 K. This transition temperature shows hysteresis associated to the nucleation and growth of ferromagnetic submicrometer clusters, that turns out to be different between cooling and warming processes. A first temperature sweep further demonstrated that the measurements were very sensitive to the phase transition and showed changes in both amplitude and resonant frequency (see fig. 1). The changes in the value of the minimum of the reflected signal $S_{11}$ from the coax resonator during the zero-field–cooled curve measured during warming (ZFC) and the field-cooled cooling (FCC) curve, together with the frequency shifts, are the consequences of the phase transitions occurring in the sample; a clear transition appears at temperatures $T_C = 60$ K on cooling and $T_C = 100$ K on warming associated to the nucleation and growth of ferromagnetic submicrometer clusters. Indeed, the blocking temperature at $T_B = 25$ K, associated to the energy barriers between the two low-temperature metastable states, is clearly seen in the ZFC curve [15,16]. In fig. 1 the temperature regions of different phases assuming a ZFC process and zero field are also depicted; mostly CO-AFM above 100 K, phase separated (PS), (i.e., coexisting phases CO-AFM and CD-FM) below 100 K and the same phase separated freezes (Fz), (i.e., the dynamics of the CD-FM are blocked) below 25 K. Even though the behaviour of these microwave measurements could be associated to the evolution of the magnetisation with temperature [16], the data can be explained with a simple model of the resistive change due to the phase transitions. When the sample is mostly dominated by the antiferromagnetic phase, it is also an insulator and it turns to be transparent to the microwaves, allowing a good resonance within the coaxial resonator and, consequently, showing a smaller reflected signal. On the other hand, when the sample is mainly constituted by micrometric ferromagnetic clusters, the resistivity of the sample decreases and the resonant frequency of the system is largely perturbed, and shifted to higher frequencies. In this case, the $Q$ value of the resonator gets worse, so the resonance broadens and the amplitude decreases (thus increasing the value of the minimum of $S_{11}$). The inset of fig. 2 shows the system resonance in both cases, CO-AFM and CD-FM. We can even associate the frequency shifts detected during the phase transition to the effective shrinkage of the resonator length as a consequence of the size of the sample when it does behave as a metal.

Figure 2 shows the changes in the reflected signal (value of $S_{11}$ and frequency of the resonance) during the metamagnetic phase transition occurring at low temperatures. The sample was first zero-field cooled to the lowest temperature ($T = 3$ K) to ensure it was almost completely in a CO-AFM state [5]. Then the external magnetic field was applied and varied at a low sweeping rate of 300 Oe/min. Both the amplitude and the frequency of the resonance change slowly with the field variation as a consequence of the slow change in the phase concentration. As the applied magnetic field further increased, ferromagnetic

Fig. 1: (Colour on-line) Temperature dependence of $S_{11}$ at $H = 10$ kOe after zero-field cooling measured upon warming (circles), and field cooling, measured upon cooling (filled circles). The value of the minimum of $S_{11}$ is shown in the upper panel, and the resonant frequency is shown in the lower panel. Between the two panels it is shown an indicative scheme of the phase ranges at zero field, frozen (Fz), phase separated (PS) and antiferromagnetic (AFM).
Fig. 2: (Colour on-line) Magnetic-field dependence of the frequency (squares) and the minimum of $S_{11}$ (triangles) for a slow sweep of the magnetic field at 3 K, with a sweeping rate of 300 Oe/min. The sample was first zero-field cooled and then a magnetic field up to 4 T was applied. After the phase transition had occurred, the magnetic field was swept back up to a negative value of $-4$ T. The inset shows the frequency dependence of the reflection coefficient $S_{11}$ at the beginning of the sweep (solid line), when the sample was in its CO-AFM phase, and at the end of the sweep (dashed line), when the sample was in its CD-FM phase.

clusters appear in the sample producing a large shift of the resonant frequency and a change in the amplitude. At $H = 40$ kOe the entire sample is in the CD-FM phase, and both the amplitude and the resonant frequency correspond the resonance of the system in this new state. In the mean time, during the phase transition, we observe a depolarization peak, characterized by an increase in the losses of the resonator that is observed through a broadening of the resonance peak together with an increase of its amplitude. Once the phase transition is completed, the magnetic field dependence of the reflected signal from the resonator remains rather constant and no hysteresis is detected. The inset of fig. 2 shows the spectra of the resonant peaks, $S_{11}$, for both the initial state after the ZFC where the sample is dominated by the CO-AFM phase, and the final state, when after the transition, the sample is completely in the CD-FM phase.

In order to have phase transition avalanches, a faster sweep of the external magnetic field is required [5]. The results of these experiments are shown in fig. 3. The sample was first zero-field cooled to the desired temperature in order to have the sample in its CO-AFM state. Then the field was varied at about 300 Oe/s. In this figure we show the results obtained at two temperatures, $T = 8$ K and $T = 3$ K. At $T = 8$ K (fig. 3(a)) the phase transition occurs smoothly over a field range of about 0.5 T, corresponding to a time window of 20 seconds. On the other hand, when the temperature was decreased to $T = 3$ K, the phase transition between the CO-AFM and the CD-FM phases occurs in less than one millisecond (see fig. 3(b) and inset).

This result is in full agreement with previous very recent results [5] which demonstrate that the magnetic avalanche in LPCMO, associated to the motion and growing of the CD-FM clusters throughout the entire sample, is of the order of few milliseconds. Resistivity measurements performed with 4 probe method [8] clearly showed that the percolative steps and, particularly, the CMR jump, corresponding to the avalanche, took only few hundreds of microseconds. The inset of fig. 3(b) shows the evolution of the reflected microwave signal $S_{11}$ during an avalanche and the time turns out to be around 500 µs. To measure the accurate curve of the inset of fig. 3(b) the frequency has been locked at $f = 3.88$ GHz in order to increase the time resolution.

High frequency. – The second set of experiments was performed at higher frequencies using cavities. A millimetre vector network analyser, MVNA-8-350 [17] was used, as a source and detector, to monitor the amplitude and phase of millimetre-wave radiation transmitted through a resonant cavity. Our cavity was an enclosed cylindrical resonator, with a diameter of 10 mm and 8 mm long, that was able to be inserted into a high-field magnet cryostat. It should then be pointed out that because, in this case, we measured transmission, the behaviour of the amplitude of the resonance is essentially opposite to that of the previous experiment. In fact, the data compare extremely well if one inverts the amplitude from the first set of measurements.
The location of the transition, and the amplitude increases again after the transition, though it does not recover the original amplitude. In fact, before sweeping the field, the $Q$ value of the cavity is $\sim 20000$ due to the transparency of the sample. At the very moment the phase transition occurs, the two phases (CO-AFM and CD-FM) compete and, thus, the cavity resonance is practically destroyed due to significant AC losses in the sample. When the transition to the CD-FM phase is completed, the cavity resonance is again recovered (see inset of fig. 4) due to the increased conductivity of the sample. However, the $Q$ value ($\sim 15000$) does not recover the value associated with the CO-AFM phase, indicating that the sample remains somewhat lossy. The phase lock maintained the tuning of the cavity. Thus, the loss peak indicates the location of the transition. Even though the phase transition is clearly observed at these higher frequencies with qualitatively the same features seen at low frequencies (see fig. 3(a)), no avalanches were detected for any field sweep rate in this case, supporting the idea that the sample should exceed a certain size in order to induce avalanches [7,18].

Conclusion. – We have reported a new method to study and detect phase transitions involving colossal changes in resistivity with microwaves techniques. In this paper we have shown the enormous importance of the microwave absorption when dealing with studies of the first-order phase transitions in manganites. Moreover, we have shown that using these techniques it is very easy to follow both, the slow and the very rapid variations associated to phase dynamics. The results also show independence from the frequency of work (low and high frequency).

FM thanks the Spanish Ministerio de Educació y Ciencia for a research grant. JMH thanks the Ministerio de Educación y Ciencia and the Universitat de Barcelona for a Ramón y Cajal research contract. ND and GA thank the European project NANOSPIN for research contracts and funding. SH acknowledges the support of the US National Science Foundation (grant nos. DMR0239481 and DMR0414809).

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Fig. 4: (Colour on-line) Magnetic-field dependence of the frequency and amplitude of the transmission signal when the phase was locked. The sweep rate of the magnetic field was 500 Oe/s and the temperature $T=3\,\mathrm{K}$. The inset shows the frequency dependence of the amplitude of the cavity with the sample before and after the phase transition. The black curve corresponds to the sample in its insulating state with a cavity $Q$ factor of $\sim 20000$, while the dashed curve corresponds to the sample in its stable CD-FM state with a cavity $Q$ value of $\sim 15000$.

(see figs. 2 and 4). A sample of La$_{0.225}$Pr$_{0.4}$Ca$_{0.375}$MnO$_3$ of dimensions $0.1\,\mathrm{mm} \times 0.1\,\mathrm{mm} \times 0.1\,\mathrm{mm}$ was glued to the base of the cavity at a distance $r/2$ from the center. The TE$_{011}$ mode, with $f = 51.875\,\mathrm{GHz}$, was excited having $Q$ values around 20000.

Results. Temperature dependence of the resonant frequencies showed a very similar behaviour to that we reached with the coaxial resonator at lower frequency. In general, two stages have been detected, corresponding to the two different phases. In the inset of fig. 4 we can see the spectra of the transmission signal when both the sample was mainly in the CO-AFM phase ($T=3\,\mathrm{K}, H=0$) and the sample was entirely in the CD-FM, ($T=3\,\mathrm{K}, H=40\,\mathrm{kOe}$). When the magnetic field was removed, the spectrum of the transmitted remained unchanged; indeed, no dependence on the magnetic field was observed, emphasising the fact that the cavity response is basically driven by the different resistivity of the sample in the two phases.

Figure 4 shows the amplitude and frequency of the transmission signal at $T=3\,\mathrm{K}$ as a function of the applied magnetic field. Experiments were performed in the phase lock mode to maintain tuning of the cavity and, therefore, better detect the shift of the resonance frequency under a swept magnetic field. First, the sample was zero-field cooled in order to obtain a basically pure CO-AFM (insulating) state. Then the applied magnetic field was swept to 40 kOe at 500 Oe/s. The graph clearly shows a depolarization peak [13] in the amplitude at around $H=30\,\mathrm{kOe}$. The loss peak (dip in this case) is seen at the location of the transition, and the amplitude increases...
Microwave detection on manganites


