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Low-temperature glassy response of ultrathin $La_{0.8}Ca_{0.2}MnO_3$ films to electric and magnetic fields

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The glassy response of ultrathin films of $La_{0.8}Ca_{0.2}MnO_3$ in the mixed phase to external magnetic and gated electro-static fields have been studied at low temperatures. The response of the resistance to external fields provides direct evidence for a hierarchical energy landscape, with strong cross-couplings between spin and charge. Magnetic coercivity measurements indicate that strong magnetic disorder accompanies the mixed phase in these films. This magnetic disorder, and the resultant coercivity, can be decreased by cooling in a large magnetic field or by electrostatic gating.

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Manganites, known for their "colossal" magnetoresistance (CMR), possess a rich diversity of phases driven by correlations between the spin, charge, and orbital degrees of freedom of the electrons and their strong coupling to the lattice.[1] Localized electrons on Mn^{3+} sites[2] create lattice distortions via Jahn-Teller-like effects, causing strong strain fields to develop. When these electrons get delocalized, for example by double exchange between aligned Mn spins, the local strain is relieved. Under appropriate circumstances, manganites have an admixture of phases of different electronic, magnetic, and structural properties, but of nearly equal free energies.[3] Consequently, the properties of these systems may be susceptible to external perturbations that lead to phase conversion within the admixture,[4] giving rise to "colossal" effects. This conversion involves rearrangement of many coupled degrees of freedom spanning all relevant length scales. The presence of competing strain fields, Coulomb interactions, magnetic correlations, and disorder may frustrate this process, giving rise to a complex free energy landscape with many nearly degenerate minima and hierarchical barriers. This naturally gives rise to glassy dynamics,[5,6] at low temperatures. Our understanding of the system's response to external forces is complicated by the cross-couplings between the different degrees of freedom. However, cross-couplings present the opportunity to influence one kind of order, e.g., magnetization, with a force that couples to a different variable, e.g., a gate electric field that couples to charge.

In this report we investigate the response of ultrathin $La_{0.8}Ca_{0.2}MnO_3$ (LCMO) films to electrostatic and magnetic fields. This composition is close to the phase boundary between a ferromagnetic metal (FM) at higher Ca doping and a ferromagnetic charge ordered insulator (F-COI) at lower doping. Single crystals of similar composition are believed to exist in a mixed phase with coexisting insulating and metallic regions,[7] with the transport properties at low temperatures arising from percolation of metallic regions. The samples are typically 21 u.c. (~82 Å) LCMO films grown using ozone-assisted molecular beam epitaxy (MBE) on surface treated $SrTiO_3$, thinned to 35–50 μm locally,[8] permitting a field-effect geometry. A 1000 Å Pt electrode deposited on the back of this thinned region is the gate. The as-grown manganite film is patterned into a wire 100 μm wide, with tabs for carrying out four terminal measurements. The gate-drain current was monitored and remained below 0.6 nA, while the source-drain measurement current was 100 nA.

We observe a magnetic transition at about 150 K, and an accompanying resistive transition, from an activated insulating state to a nominally metallic state near the Curie temperature, along with CMR. However, below 36 K there is a reentrant insulating phase.[9] Near the resulting minimum, the resistance has a large susceptibility to gate and magnetic fields, with clear signatures of glassy dynamics and hierarchical energy barriers. We argue that the dynamics are governed by a variable that is cross-coupled to both charge and spin degrees of freedom. Cross-couplings have been exploited to change the sample's magnetic coercivity upon application of a gate electric field. The details of the gate effect at low temperatures have been discussed elsewhere.[10] For the present purpose, it suffices to note that the gate electric field couples to the charge degrees of freedom, while the magnetic field couples to spin. The applied gate voltage is always negative, inducing "hole"-like charge carriers.

Within a mixed phase scenario, measurements of resistance are particularly sensitive to changes in the percolative metallic path. This is especially true for our ultrathin films. Upon applying an external field that favors the growth of one phase with respect to the other, the change in resistance depends on the motion of domain boundaries separating the two. In this context, the presence of hierarchical energy barriers was demonstrated[11] in resistivity measurements of $La_{0.5}Ca_{0.5}Mn_{0.95}Fe_{0.05}O_3$ in magnetic fields. We have measured a similar hierarchical response in our films to both applied electric and magnetic fields at 30 K. A succession of external magnetic/gate electric field pulses of different values and duration were turned on and off for times of 0.5–1.5 h, respectively. When a field was turned on, the resistance decreased to a value R_{ON}, with a large "fast" change and a smaller "slow" part that evolved with a logarithmic dependence on time [Figs. 1(a) and 1(b)]. On turning off the field, the resistance relaxed to an intermediate value RTR (thermo-remnant resistance), indicating that the sample resistance undergoes an irreversible change, analogous to thermo-remnant magnetization in spin glasses.[12] The fast part of this recovery is repeatable, but the slow part shows signs of aging, i.e., it depends on how long the field was on for. On subsequent pulses, if the value of the field exceeded that of the maximum field previously applied, R_{TR} decreased further, but otherwise the response was nearly reversible [Figs. 1(c) and 1(d)]. Thus, the R_{TR} has memory of the previous highest field applied. Furthermore, the R_{TR} does not discriminate between the application of an electric or magnetic field. This was demonstrated by first turning on a large gate electric field, turning it off, and following that by a series of increasing magnetic field pulses [Fig. 1(e)]. The hierarchy of barriers for the lowering of resistance due to an applied magnetic field seems to "respect" the barriers already crossed by application of the gate electric field, regardless that they couple to spin and charge, respectively.

We interpret these results in terms of the dynamics of interphase domain walls (IDWs) that separate the insulating and metallic regions in the mixed phase, in a hierarchical pinning landscape. The application of an external field lowers the free energy of one phase with respect to the other, and the IDWs feel an effective force. Pinning sites up to a certain strength are then overcome and effectively eliminated, and the walls move irreversibly into a new configuration. For subsequent fields of lower strength, the elasticity of the IDWs allows them to respond in a reversible manner to an on/off sequence because of the relatively pinning-free landscape. When an external field greater than the previous highest field is applied, the next level of pinning sites are overcome, and another irreversible change occurs. The mutually respected hierarchical barriers in the response of the resistance to electric and magnetic fields suggests that this energy landscape is a function of a variable that is cross-coupled to both spin and charge. Strain is a possible candidate, where competition between long-range strain fields and local disorder can cause frustration[13] and pinning of domain walls.[14] In this scenario, it does not matter that frustration is relieved by aligning spins or inducing charge at the domain walls, since they cross-couple to the same strain field.

We now turn to the magnetic coercivity of our ultrathin films. We use an unpatterned film (21 u.c.) with a 12 mm^2 gate to detect small changes in the film's magnetic moment using a SQUID magnetometer. The coercivity H_C at 2 K was 484 Oe, significantly higher than that of thicker films, and is also enhanced from its value at 70 K of 147 Oe. This is believed to be due to the pinning of magnetic domain walls at defects caused by magnetic inhomogeneities[16] and has been observed in other studies on manganite thin films.[15] Furthermore, upon cooling the film in a 5.5 T field from above T_c to 2 K, H_C was reduced by about 120 Oe compared to the zero field cooled value. Thus, in a more metallic or homogeneous phase, the pinning is weakened, implying that the magnetic inhomogeneities arise from the mixed phase.

Further evidence of magnetic disorder is found in the observation of return point memory (RPM) [Fig. 2(a)] in the hysteresis loops. This implies that the magnetic disorder in the mixed phase gives rise to a spread in the local coercivity, as is required in models postulated by Sethna *et al.*[17] of interacting Ising spins in the presence of magnetic disorder for observation of microscopic RPM. Our observations also imply the absence of antiferromagnetic interactions between

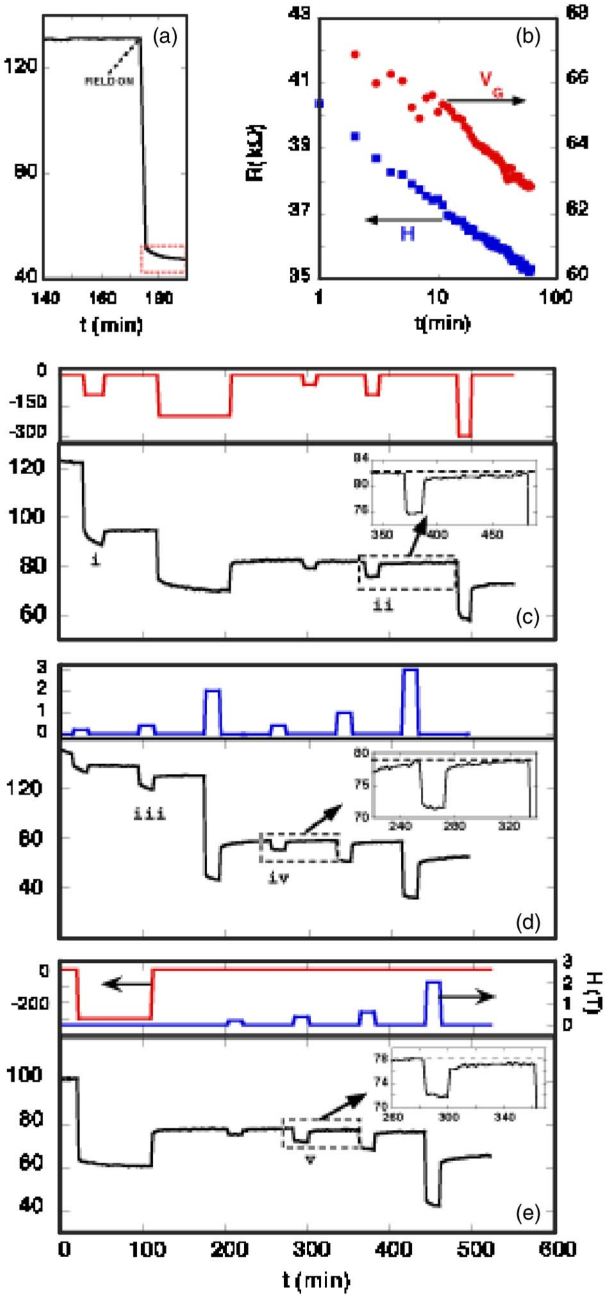

FIG. 1. (Color online) Glassy response: (a) Upon turning on a field, a large fast change in resistance is followed by a smaller glassy change. (b) The response $R(t)$ after turning on a magnetic field of 2.5 T (200 Oe/s) and gate voltage of -300 V (85 kV/cm), after cooling in zero field to 30 K. Units are resistance in $k\Omega$ for both vertical axes. (c) Upper panel shows gate voltages, sequence of -100, -200, -50, -100, and -300 V with intervening zeros, and lower panel shows the corresponding changes in resistance. (d) Sequence of magnetic fields of $H=0.2$, 0.4, 2, 0.4, 1.0, and 3.0 T with intervening zeros and response. (e) Equivalence of barriers: gate voltage pulse of -300 V followed by magnetic field pulses of 0.2, 0.4, 0.6, and 2 T with intervening zeros. The insets show detail of recovery in a nearly reversible manner. For relevant comparisons, (*i*) and (*ii*) are responses to the equal pulse sizes in volts; (*iii*) and (*iv*) are for equal pulse sizes in H; (*v*) is response to same H pulse as in (*iii*) and (*iv*).

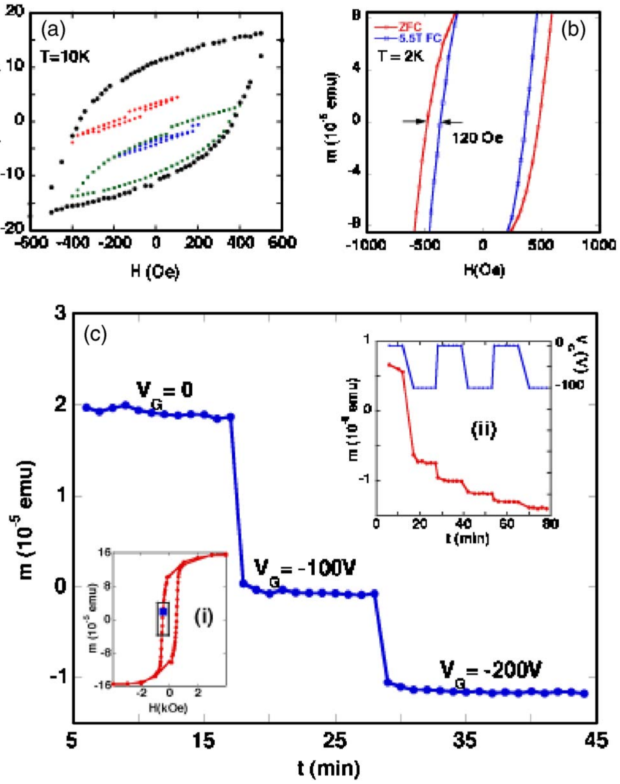

FIG. 2. (Color online) (a) Return point memory measurements at 10 K, with hierarchical loops. (b) Lowering of H_C upon field cooling in 5.5 T, to 2K. (c) Gate-induced change in magnetic moment. A magnetic field of −450 Oe was applied at $t=0$ after saturating at 5000 Oe. The gate voltage was changed in two steps of 100 V each. Inset (*i*) indicates section of hysteresis loop where the gate was turned on. Inset (*ii*) shows the response upon repeated on/off cycling of a gate voltage of −100 V, with the sample state prepared in the same manner as in the main figure (c), but with the field biased at −475 Oe.

spins, since these lead to a violation of the "no pass" criterion, a necessary condition for RPM.[18] Within the Sethna model, at H_C there is an infinite avalanche, where a macroscopic number of spins in the sample flip due to interactions between spins. However, for sufficient magnetic disorder, this condition is never obtained, and the spins flip in a hierarchy of coercive fields, as observed in our sample. At low temperatures, we observe very slow glass-like dynamics in the evolution of the magnetic moment when we bias the magnetic field at a value close to the nominal coercive field (half-width in H near $M=0$), consistent with a picture of thermally assisted motion of magnetic domain walls (MDWs) across hierarchical barriers.

Much interest exists in controlling or altering magnetic behavior using electrical means, such as injection of spin-polarized current[19] in metallic systems and the application of a gate voltage[20] in dilute magnetic semiconductors. Motivated by the strong cross-couplings observed in the resistance response of our films, we measured the effect of a gate electric field on the magnetic moment of a sample biased near H_C . The sample was cooled in zero field and then saturated by applying +5000 Oe. The field was then reversed to −450 Oe, a gate voltage of −200 V was applied in 100 V steps, and the moment was measured as a function of time [Fig. 2(b)]. Upon application of the gate voltage, the sign of the total moment is reversed in this case, indicating that the nominal H_C was crossed. Considering that the effective gated area was only about 33% of the total film area, the change in magnetic moment of the gated area was about 50% of the saturation value. Furthermore, upon turning off the gate electric field, we observe the sample magnetic moment changes by a small amount in the *same* direction as before, i.e., in the direction of the applied magnetic field. We repeated our measurements, this time cycling of the gate electric field in on-off sequences. We observed that the sample magnetic moment keeps changing [Fig. 2(b) inset] in ever smaller steps at each pmpff edge of the voltage pulse. Thus, there is a ratchet-like effect that allows change in only one direction, and repeated application presumably makes the sample's magnetic moment converge towards a "stable" state.

We interpret these changes in coercivity upon gating in terms of the motion of MDWs through a random pinning potential[21] caused by magnetic inhomogeneities. Particularly, we consider a scenario where pinning sites lower the energy of the MDWs (equivalent arguments may be constructed for the converse case). Starting with all spins in one direction (up) and reversing the magnetic field, down-spin domains are nucleated and these domains grow at the expense of the up-spins to lower the Zeeman energy, but pinning of MDWs impedes this process. Furthermore, the MDWs cost energy, and this provides an effective elastic modulus for increases in their length as they configure themselves to minimize the total energy (Zeeman+elastic+pinning). Motivated by our observations of a hierarchical response of the resistance to a gate electric-field, we present an analogous scenario for the response of the MDW pinning landscape to gate pulses. When the sample is biased in a magnetic field close to H_C, and an initial gate electric field is applied, the metallic phase fraction increases, making the system more homogeneous as in the magnetic field-cooled case. During this first gate pulse, we presume that all magnetic pinning centers below a threshold are wiped out, while those above this threshold are weakened. During this time, MDWs are depinned, and the magnetic moment increases in the direction of the applied magnetic field, until the MDWs find a new metastable configuration. When the gate is turned off, pinning centers above the threshold that had only been weakened by the gate now reappear fully. The MDWs will now reconfigure themselves, but always prefer moving into a well that lowers the Zeeman energy. Thus, the presence of the external magnetic field provides the impetus for ratchet-like behavior, and causes further change in the moment along H. Repeated application of a gate field less than or equal to the highest previously applied field should lead to reversible modulation of the strength of the magnetic pinning centers. Thus, due to repeated ratchet action, the system settles into progressively deeper minima, until the modulation of the pinning potential caused by a given gate voltage is insufficient to cause further depinning.

In conclusion, the glassy behavior is not like that of a glass consisting purely of spin or charge, but a cross-coupled variable. If the underlying mechanism involves long-range strain fields, it would imply that the glass is of a very nonlocal nature. Ahn *et al.*[14] have shown that the combined effects of long range strain fields and local intrinsic disorder naturally give rise to multiscale mixed phase domain structures and a metastable energy landscape, which could in principle give rise to the observed hierarchical response. Furthermore, we have shown that magnetic disorder arises *intrinsically* in the mixed phase, leading to pinned MDWs, etc., and that this disorder can be tuned by external electric and magnetic fields to effect changes in magnetic behavior. While this may be of some practical interest, it is a novel system for studying the dynamics of elastic manifolds in random pinning potentials in 2D (MDW is likely thicker than film), with the unique capability of being able to tune the strength of the magnetic disorder potential with a gate.

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