

Manganite-based devices: opportunities, bottlenecks and challenges

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Since the rejuvenation of interest in the rare earth manganites owing to their potential use as magnetoresistive sensors, there has been adequate research to arrive at some evaluation of the potential for these materials in a variety of technologies that would use the peculiar properties of these materials. The magnetic field sensitivity of the transport properties, the strong metal insulator transition at the Curie temperature, the electric field polarizability of the material and its subsequent effect on the transport properties, the half metallicity of the electronic bands, etc., are properties of the rare earth manganites that could be exploited in a variety of devices. In this review we explore the various interesting technological avenues that are being pursued and address the uniqueness of the material that may enable a given technology as well as the various bottlenecks that will have to be overcome in order to successfully compete with the existing technologies. As is the case with many emerging materials technologies, the devices also serve as a vehicle for further understanding of the fundamental mechanisms behind the basic properties of the materials.

Keywords: colossal magnetoresistance; manganite; spin polarization; bolometer; spin injection; electric field effects

1. The unusual properties of rare earth manganites

While rare earth manganites have been studied several decades ago (Jonker & Santen 1950; de Gennes 1960; Watanabe 1961; Volger 1954), there has been a rejuvenation of interest in this material system over the last few years on account of several factors: the deployment of magnetoresistive sensors in commercial magnetic memory systems (Simonds 1995), the significant advances in the fabrication of epitaxial oxide films since the emergence of high temperature superconductivity, and the growing interest in transport phenomena in the oxides in general where strongly correlated electronic phenomena are manifested.

The manganites constitute a very broad spectrum of magnetic materials as a function of doping with unusual transport properties where, by varying the dopant and the temperature, one can realize ferromagnetic or paramagnetic conductors and ferromagnetic, paramagnetic, antiferromagnetic or charge ordered insulators (Schiffer *et al.* 1995). In many of these phases the transport properties are sensitive to the magnetic fields and metal–insulator transitions occur at a variety of compositions. To simplify the discussion let us focus on the $\text{Re}_{0.7}\text{M}_{0.3}\text{MnO}_3$ system (where Re = trivalent La, Nd, Pr, ... and M = divalent Ca, Ba, Sr, ...) and in particular on $\text{Nd}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$, whose R – T curve is shown in figure 1 (Xiong *et al.* 1995). The zero

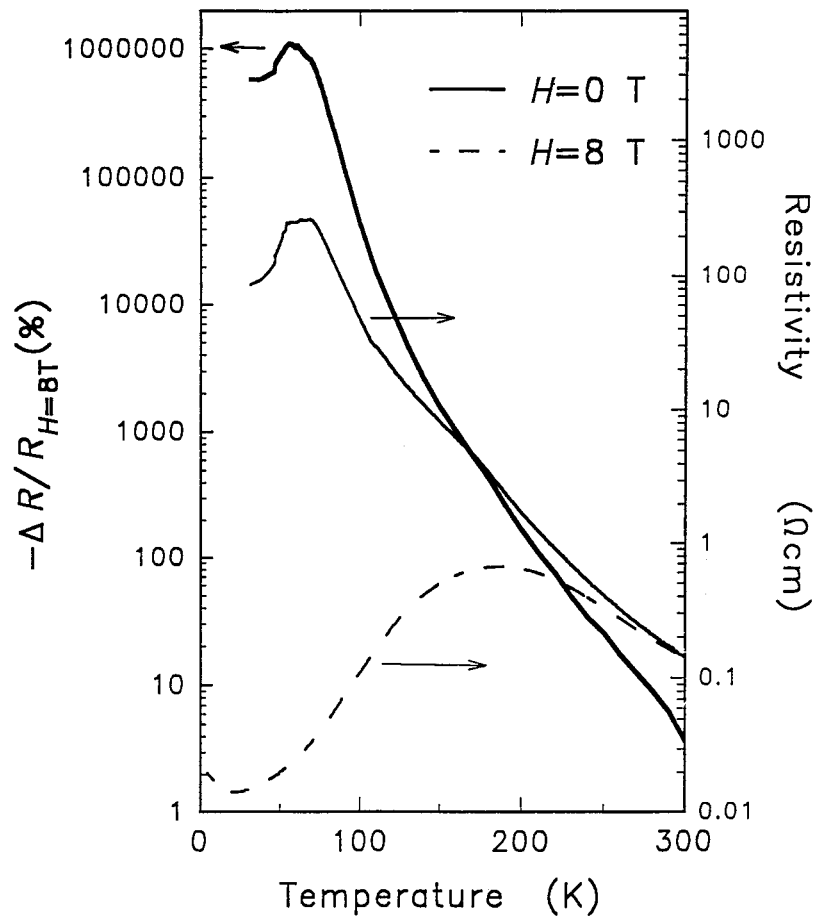


Figure 1. Resistance versus temperature curve for an epitaxial film of $\text{Nd}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ in zero and 8 T magnetic field.

field curve, an object of continuing curiosity for the theorists, consists of a paramagnetic semiconducting region above T_C with a well-defined activation energy of about 125 meV (Xiong *et al.* 1995) and below T_C a metallic ferromagnetic region (Chahara *et al.* 1993). On the application of a magnetic field, albeit a large one of 8 T, there is significant change in the resistivity which has been coined the colossal magnetoresistance (CMR) effect (Jin *et al.* 1994). This is the first technologically interesting property. If one looks at the zero field resistance curve there is a sharp drop of the resistance at the T_C and this large temperature coefficient of resistance is another technologically important feature. In addition, these features are also affected by electric fields which could potentially lead to novel devices as discussed later. An extraordinary property of the manganites is the observation of half metallic conduction bands in the system by both tunnelling (Wei *et al.* 1997) and photoemission measurements (Park *et al.* 1998) which tend to suggest that 100% polarization of electrons is possible and these materials are hence an efficient source of polarized electrons and this can be also the basis for a variety of novel devices.

2. Device approaches

Based on the properties discussed above, a number of device approaches are being explored, and we will discuss them sequentially as below.

1. Magnetic field sensors (*a*) using the CMR effect in a film, (*b*) using a spin valve structure, and (*c*) as a microwave CMR sensor.
2. Electric field effect devices (*a*) using a SrTiO₃ gate and (*b*) using a ferroelectric gate.
3. Bolometric uncooled infrared (IR) sensors using the metal–insulator transition at the Curie temperature.
4. Low temperature hybrid HTS-CMR devices (*a*) flux focused magnetic transducers and (*b*) spin polarized quasi-particle injection devices.

(*a*) Magnetic sensors

(i) Using the CMR effect in a film

The industrial requirements for a magnetic sensor can be summarized as follows.

1. Operation at room temperature and up to 100 K above room temperature.
2. At least a 20% response at a field of 100 Gauss.
3. Temperature independent CMR values over 350 ± 50 K.
4. Acceptable noise values.
5. Retention of magneto-transport properties in patterned films at dimensions approaching sub-1000 Å scales. (The current thinking is that oxide-based CMR sensors will have maximum impact only on memory systems approaching densities of 100 Gb cm^{-2} .)

At room temperature the CMR effect drops dramatically and the largest effects are seen at lower temperatures in high quality epitaxial films. Further, the low field sensitivity decreases as the film quality is enhanced, leading to the idea that defects are necessary for low field sensitivity. In fact, when polycrystalline films are used there is a significant enhancement in the low field sensitivity as shown in figure 2, and this has been identified with spin polarized electron tunnelling between grains (Hwang *et al.* 1996; Gupta *et al.* 1996; Shreekala *et al.* 1997). But even in this case the sensitivity decreases as the temperature is raised so that at room temperature there is virtually little MR effect at low fields. Ion irradiation effects (figure 3*a*) (Chen *et al.* 1996) or the use of a superlattice structure (figure 3*b*) (Kwon *et al.* 1997) cause an enhancement in the CMR effect as well as broadening the temperature response of the CMR effect as desired, but the room temperature response is poor despite all these processing steps. Tokura and coworkers have studied other phases that show very large CMR effects (100% at 0.2 T fields) but at low temperatures (*ca.* 100 K) (Kimura *et al.* 1996). Thus using a thin film approach, getting a room temperature sensor with adequate low field sensitivity seems quite unlikely at this stage.

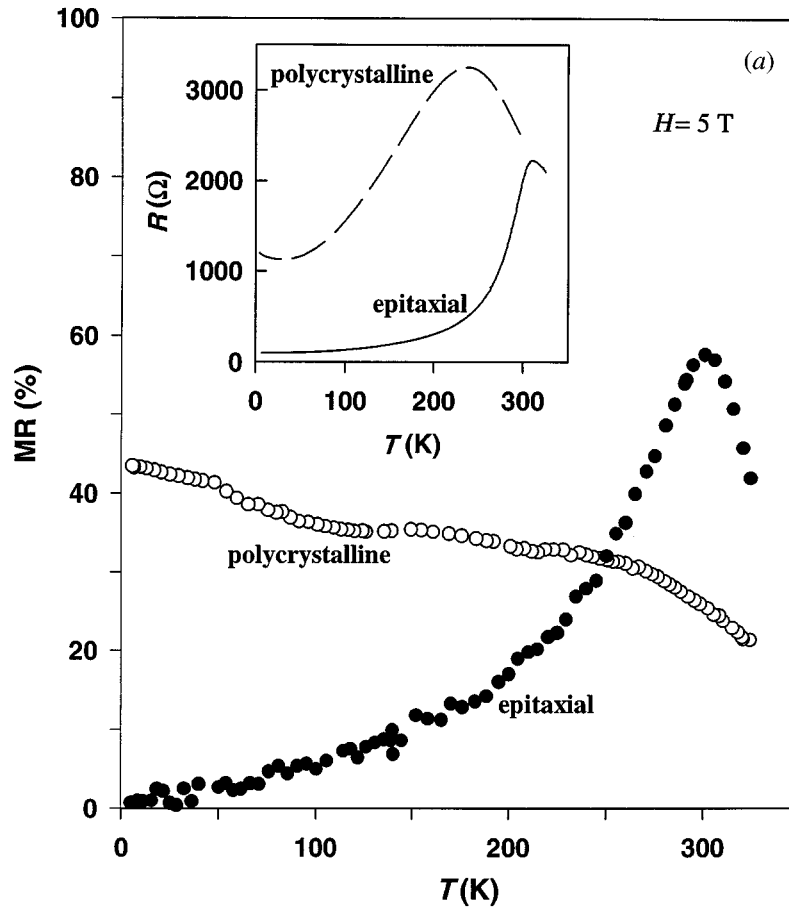


Figure 2. Resistance and magnetoresistance versus magnetic field as a function of temperature for a polycrystalline film showing: (a) high field ($H = 5$ T) response. Inset shows the comparison of the dependence of resistance on temperature for the polycrystalline and epitaxial films.

(ii) *Spin valve and spin tunnelling devices using CMR electrodes*

Spin valve devices consist of two ferromagnetic electrodes separated by a normal layer (Dieny *et al.* 1991). The lower electrode has in general a larger coercive field than the top electrode and is in general pinned via exchange biasing with a bottom antiferromagnetic layer. The entire device structure can be an epitaxial multilayer consisting of CMR ferromagnetic electrodes with a conducting layer of another epitaxial metal oxide such as LaNiO_3 (LNO), LaSrCoO_3 (LSCO), SrRuO_3 (SRO), or even YBCO. Figure 4 shows the X-ray diffraction peaks from a trilayer consisting of LSMO ferromagnetic top and bottom electrodes and an intermediate layer of LSCO (Robson *et al.* 1998). High quality epitaxial heterostructures can indeed be generated, but on devices prepared to date there is a large series resistance not explained on the basis of the resistivities of the individual layers which tends to reduce the observed magnetoresistance.

An alternative approach is the trilayer spin tunnelling device where the intermedi-

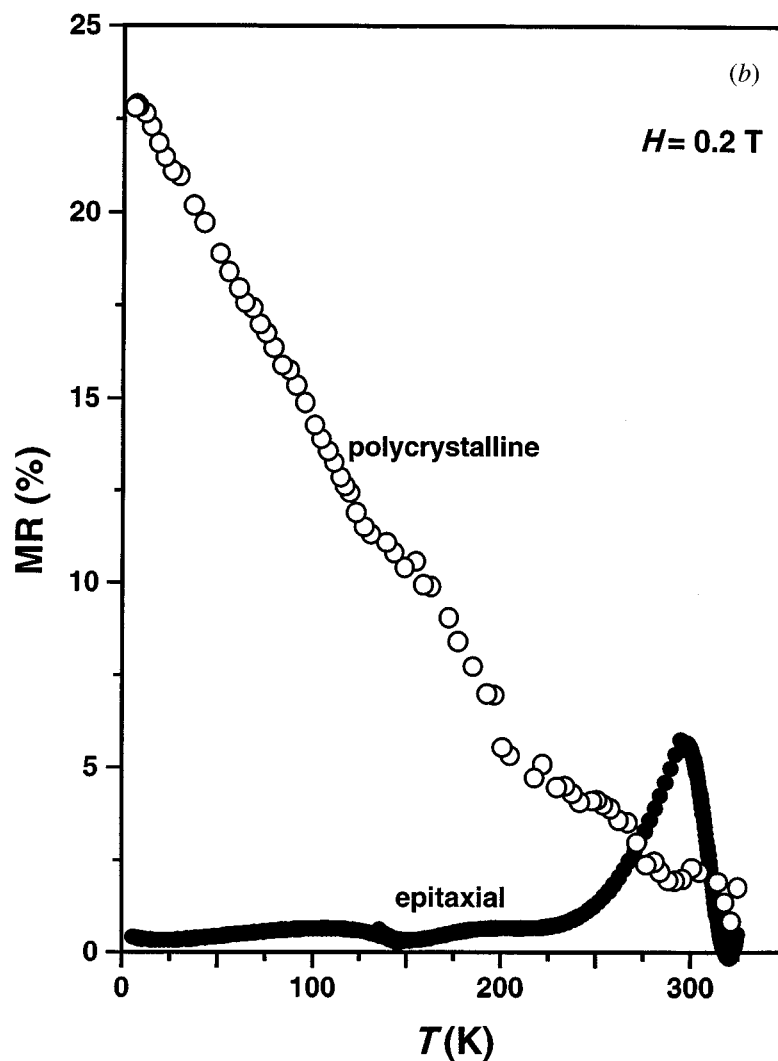


Figure 2. (Cont.) (b) the large low field ($H = 0.2 \text{ T}$) response.

ate layer is an epitaxial insulator such as SrTiO_3 (STO), LaAlO_3 (LAO), or MgO . In the first of such devices demonstrated by Sun *et al.* (1996), a very large magnetoresistance effect was indeed seen at low fields and at low temperatures which deteriorated as the temperature was raised. One of the speculations for the decrease of the magnetoresistance was the temperature-dependent quality of the STO. However, recent experimental data on the measurement of the half metallicity of the CMR material, LSMO epitaxial films to be precise, by spin polarized photoemission measurements (Park *et al.* 1998), show that while at low temperatures (less than 40 K) the minority spin subband has no density of states at the Fermi level; as the temperature is increased the two subbands overlap more and more with the polarization going to zero at the Curie temperature. This clearly suggests that unless a very high Curie temperature manganite compound is discovered, their use in such spin tunnelling or

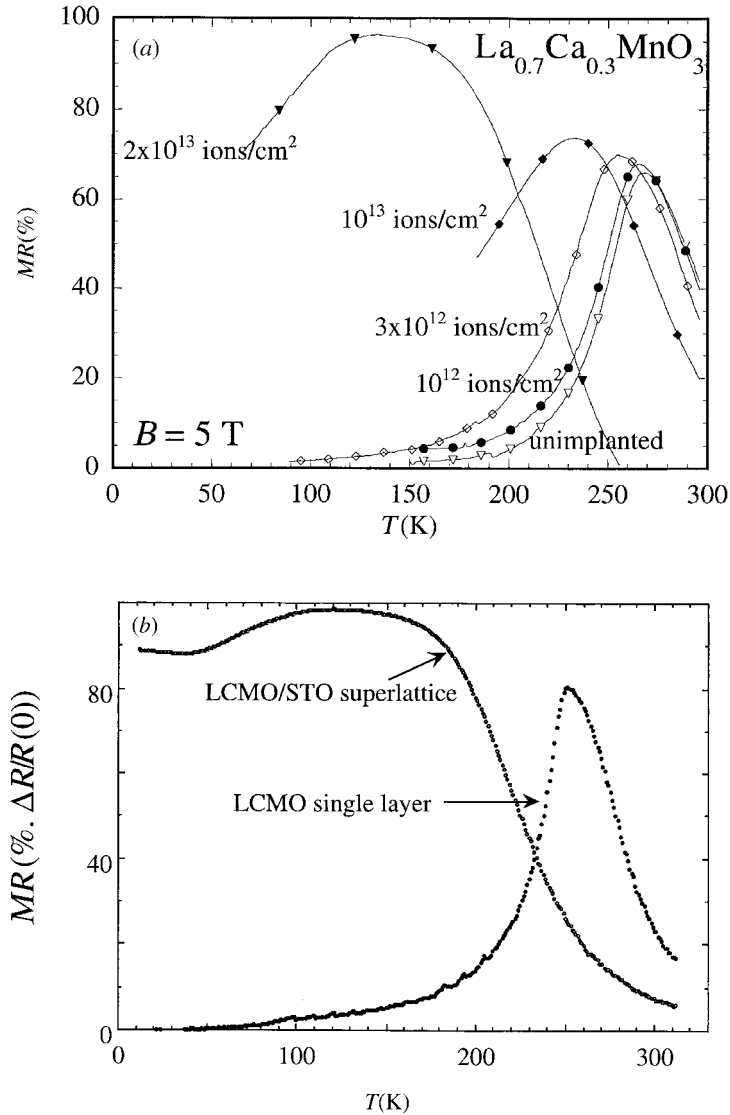


Figure 3. (a) The magnetoresistance as a function of 200 keV Ar ion irradiation dose in films of LCMO. (b) The magnetoresistance in a superlattice films of LCMO–STO (64–160 Å) in comparison with single layer of LCMO.

spin valve devices may be limited to lower temperatures. Recently, we have prepared epitaxial films of the magnetites Fe_3O_4 by pulsed laser deposition (PLD) and these films with resistivities in the milliohm range show 100% polarization even at 300 K on account of their very large Curie temperatures (840 °C). Figure 5 shows XRD data for an epitaxial trilayer spin tunnelling structure consisting of two Fe_3O_4 electrodes with an intermediate layer of STO (Ogale *et al.* 1998). In the absence of a higher Curie temperature manganite, our effort on spin valve and tunnelling devices will focus on Fe_3O_4 ferromagnetic electrodes.

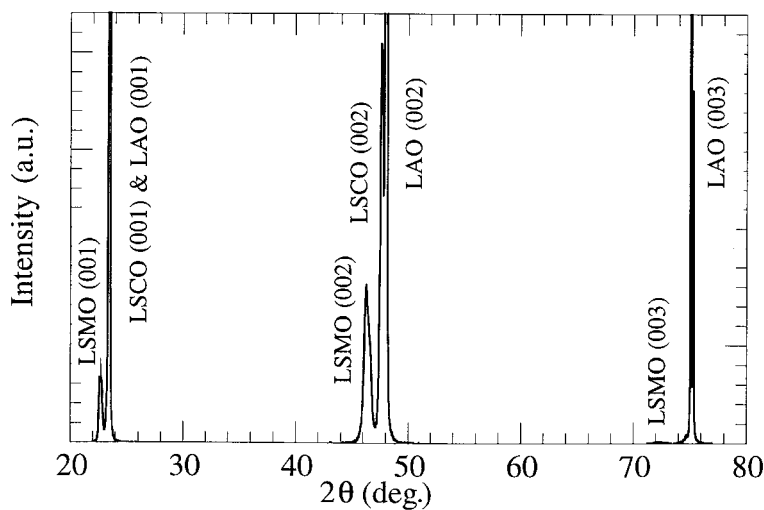


Figure 4. X-ray diffraction spectrum of a trilayer of LSMO–LSCO–LSMO showing well-oriented (100) peaks.

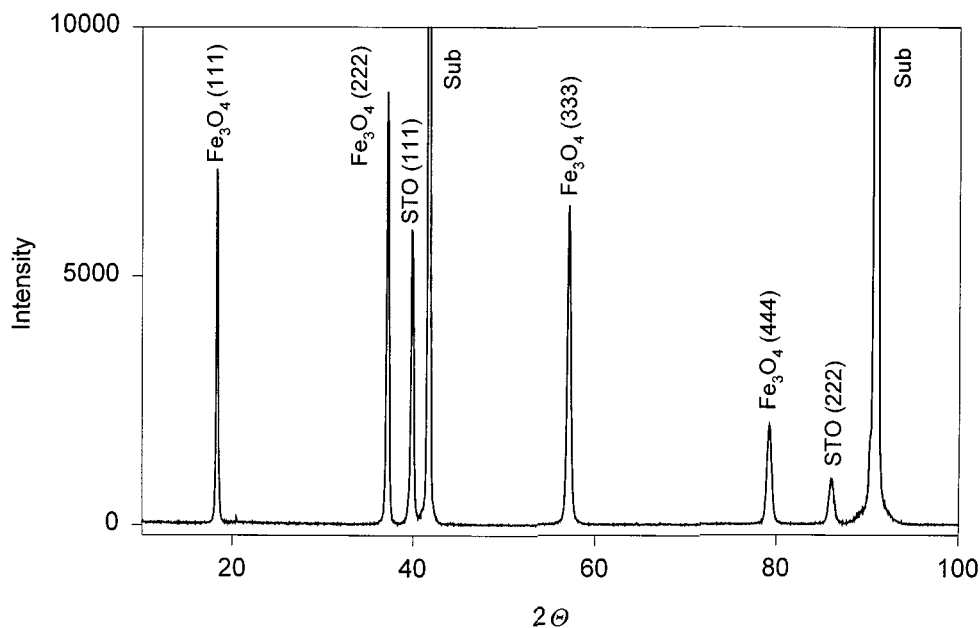


Figure 5. X-ray diffraction spectrum of a trilayer of Fe_3O_4 –STO– Fe_3O_4 showing well-orientated (100) peaks.

(iii) Microwave CMR sensor

At microwave frequencies, significant CMR effects comparable to the low frequency values have been seen (Dominguez *et al.* 1995). In fact, operating a cavity in the antiresonant mode, a very large CMR effect was seen (an effect of more than 50%) at fields of a few tens of millitesla (Lofland 1996), which suggests the possibility

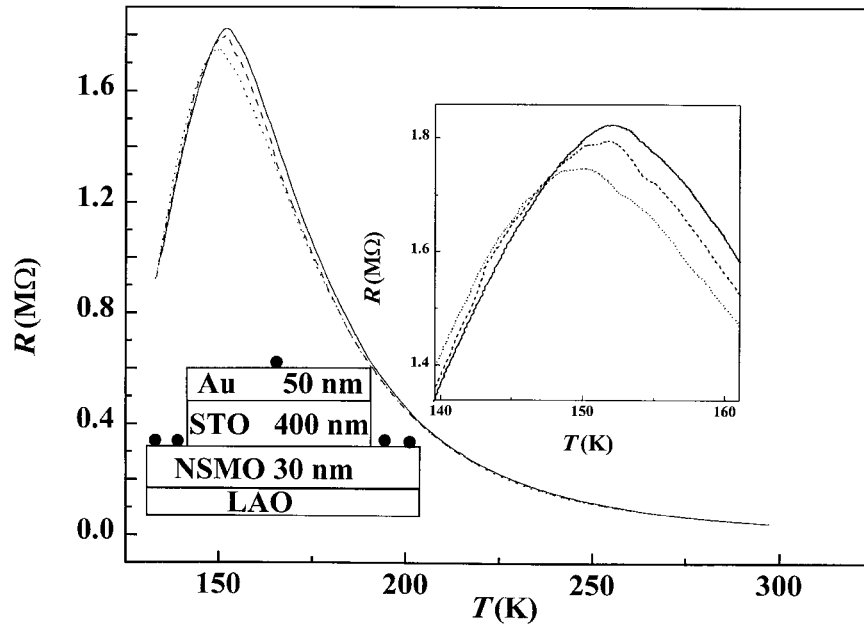


Figure 6. Electric field effect on the resistance versus temperature curve in an FET structure with a CMR channel and an STO gate; zero field (solid line), +2 V (dotted line) and -1.5 V (dashed line).

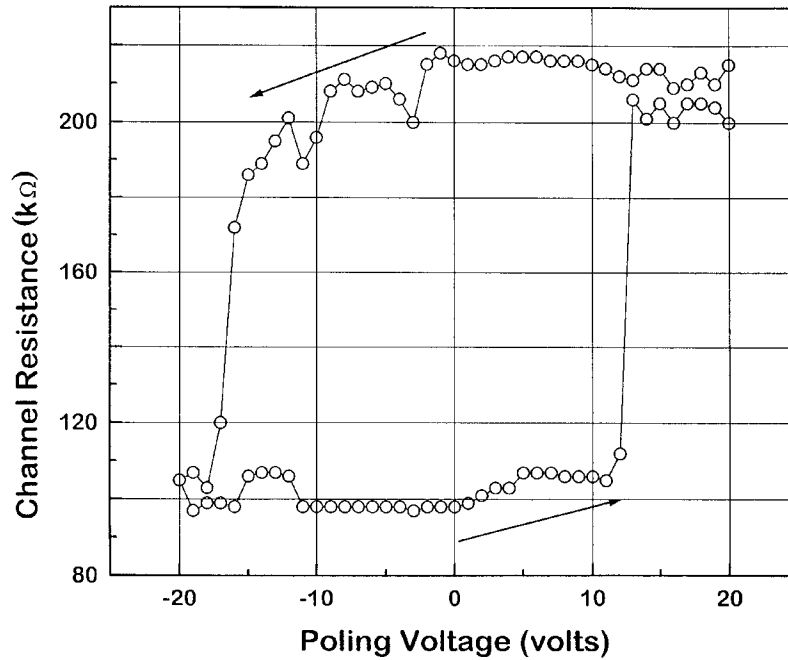


Figure 7. Resistance modulation as a function of gate voltage in an FET configuration with a CMR channel and a ferro electric gate dielectric.

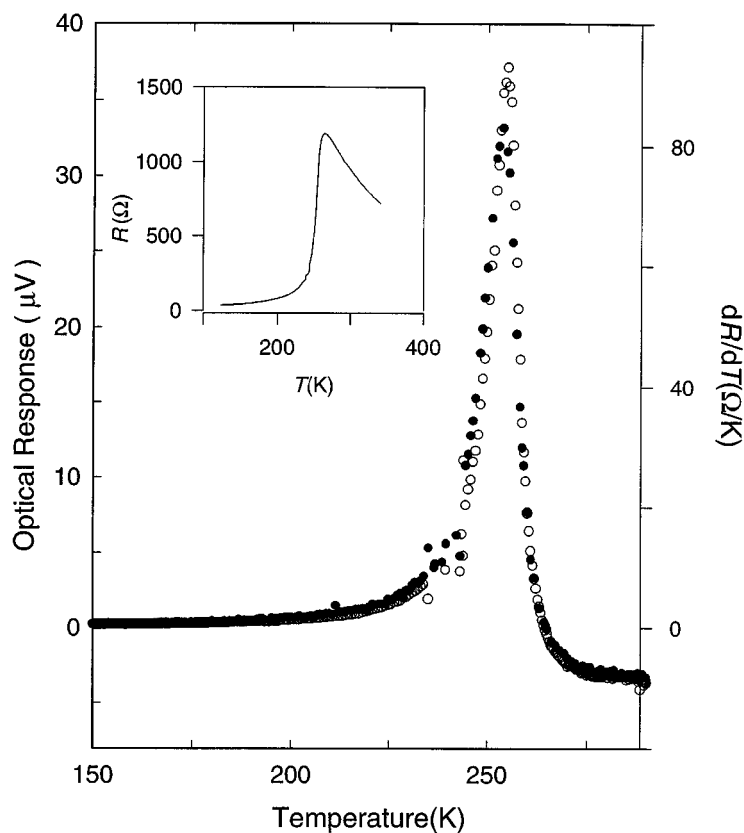


Figure 8. Optical response of a film of LCMO (closed circles) in comparison with the TCR (open circles). The R - T curve is shown in the inset.

of magnetically modulated microwave devices. Whether the dielectric losses in the material would permit usable devices, and the magnetic field levels can be further reduced are issues that require further study.

(b) *Electric field effect devices*

Non-volatile FE switches: FETs based on CMR channels show some interesting characteristics depending on the dielectric layer on top as to whether it is a paraelectric layer, such as STO, or a ferroelectric layer, such as PZT. In the case of STO, the response of the device to both polarities is the same, a general reduction in the resistance and a seeming shift of the peak resistance temperature to lower temperatures (figure 6) (Ogale *et al.* 1996). The lack of polarity effects suggests effects which are induced by polarization rather than charge injection, and the polarizability of the bonds in the solid and its subsequent effect on the transport properties seems to be the likely scenario for the mechanism behind the field effects seen. The effect is rather small at the percentage level though this technique may shed more light on the nature of the unusual transport in these materials. For example, one can learn about the dynamics of the polarization response which exhibits an unusual slowing

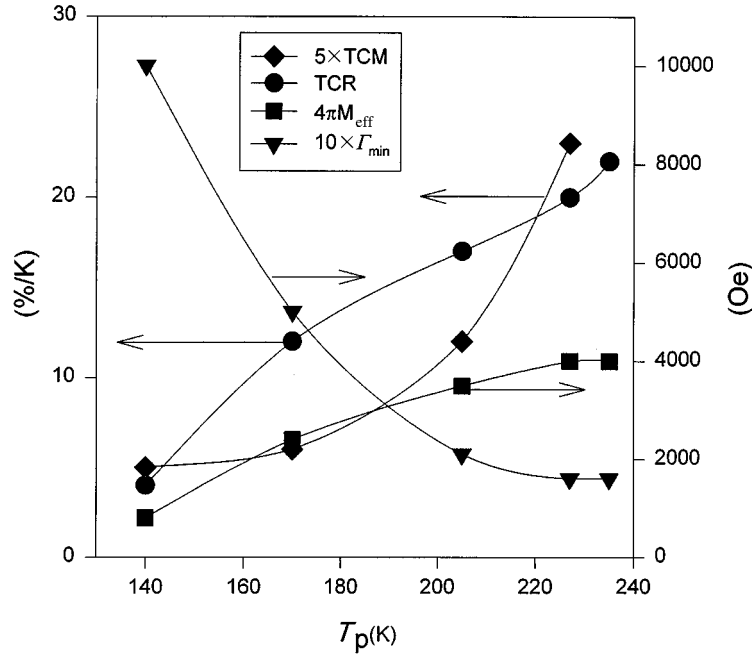


Figure 9. Correlation of TCR, TCM, magnetization and FMR line width.

down as one approaches the Curie temperature (Ogale *et al.* 1996). However, devices with a ferroelectric gate structure show significant changes in the resistance modulation as large as 100s of per cent (figure 7) (Mathews *et al.* 1997), which make them attractive for potential non-volatile device applications. The advantage of these devices unlike conventional NVRAM would be that the reading of the state of the memory would be direct since the resistances are considerably different in the two states and thus will not require a rewrite of the memory. Why these two types of devices, STO versus PZT gate, behave so differently is an issue that needs further study.

(c) Room temperature bolometric IR sensors

Due to the advances in thermoelectric cooling, materials with high thermal nonlinearities in the temperature range of 250–300 K, are potential candidates for bolometric sensors. In figure 8 is shown the optical response observed in LCMO as a function of temperature and the response correlates very well with the temperature derivative of the resistance indicating the bolometric nature of the response (Rajeswari *et al.* 1996a). The figure of merit for a bolometric material is the thermal coefficient of resistance (TCR) and the noise volume (S). The commercial bolometers based on VO_x today use TCR values around 2.5–4%. In comparison TCR values ranging from 8 to 18% are possible in the manganites LCMO over the very same temperature range. The TCR is very closely related to the magnetic homogeneity of the material and as a matter of fact the data in figure 9 shows the strong correlation between TCR and the thermal coefficient of the magnetization (TCM), the inverse ferromagnetic resonance (FMR) linewidth and the value of the total magnetization

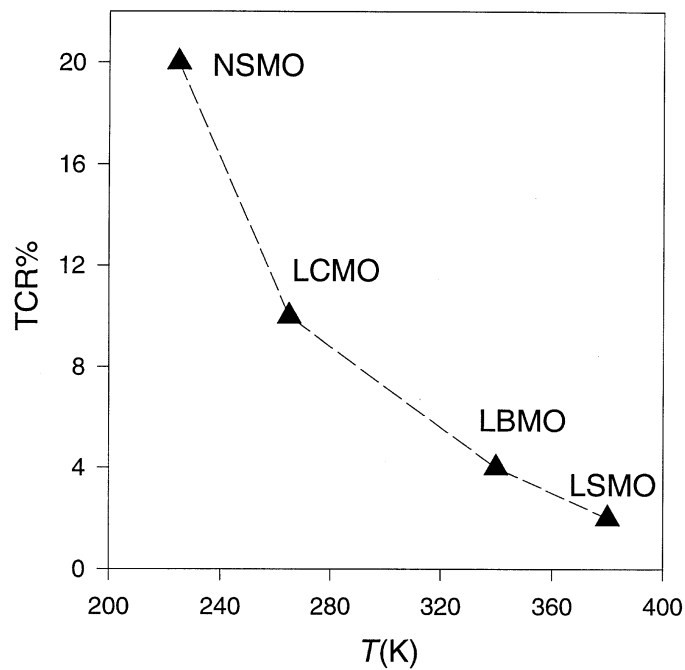
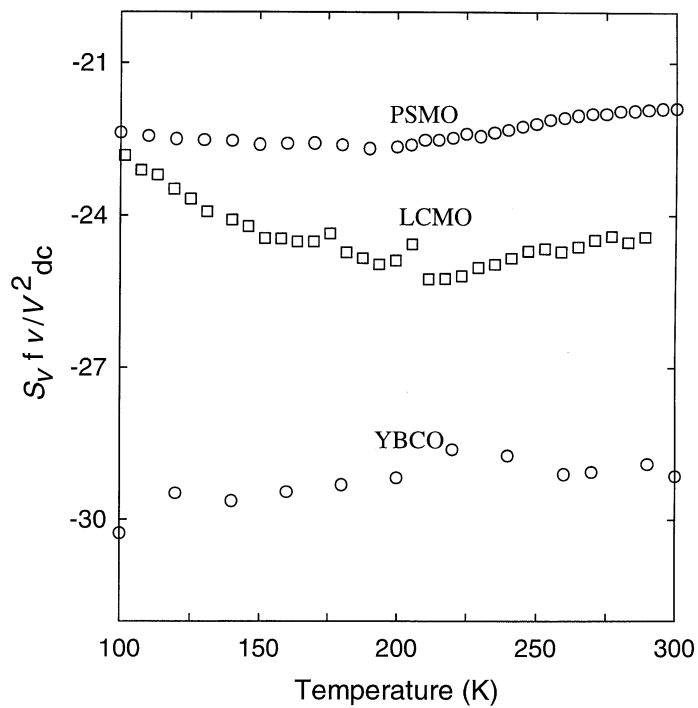
Figure 10. Maximum TCR value versus T_C .

Figure 11. Noise characteristics of some of the earlier films of manganites with respect to YBCO.

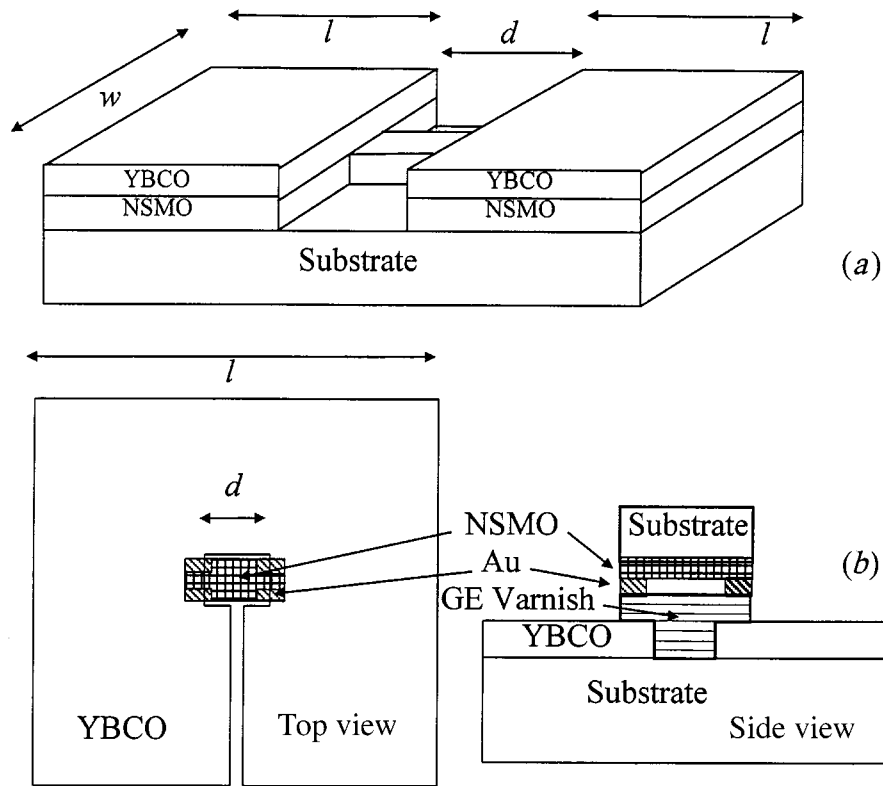


Figure 12. A schematic view of (a) gap-device and (b) washer-device. The typical size of YBCO is $l = 3$ mm, $w = 5$ mm and $d = 25$ μm or 120 μm for the gap-devices and $l = 10$ mm and $d = 1$ mm for washer-devices, where the slit width is 10 μm . The NSMO bridge in the gap device has length d and width 1 mm. (c) A side view of magnetic field patterns for both gap- and washer-devices having a flux focusing in the gap or hole, respectively, in the presence of a uniform perpendicular applied magnetic field.

(Rajeswari *et al.* 1998). The TCR value drops with the increase in the Curie temperature (figure 10) which is an indication of an intrinsic thermal inhomogeneity in these manganites (Goyal *et al.* 1997).

Early noise measurements in the manganites also showed very large noise volumes in those materials which were typically four to six orders of magnitude larger than was seen in other metal oxide conductors such as YBCO (Rajeswari *et al.* 1996b; Alers *et al.* 1996). However, recently, we are seeing significantly reduced noise volumes in higher quality manganite films and there is a clear correlation with increased TCR values. In figure 11 is shown the noise characteristics of CMR films made about a year ago in comparison with other oxide conductors such as YBCO. The films made today with much higher TCR values also show two orders of magnitude reduction in the noise volumes (Rajeswari *et al.* 1998). The noise figures in the manganite films today as well as the TCR values are very attractive for the fabrication of bolometric detectors.

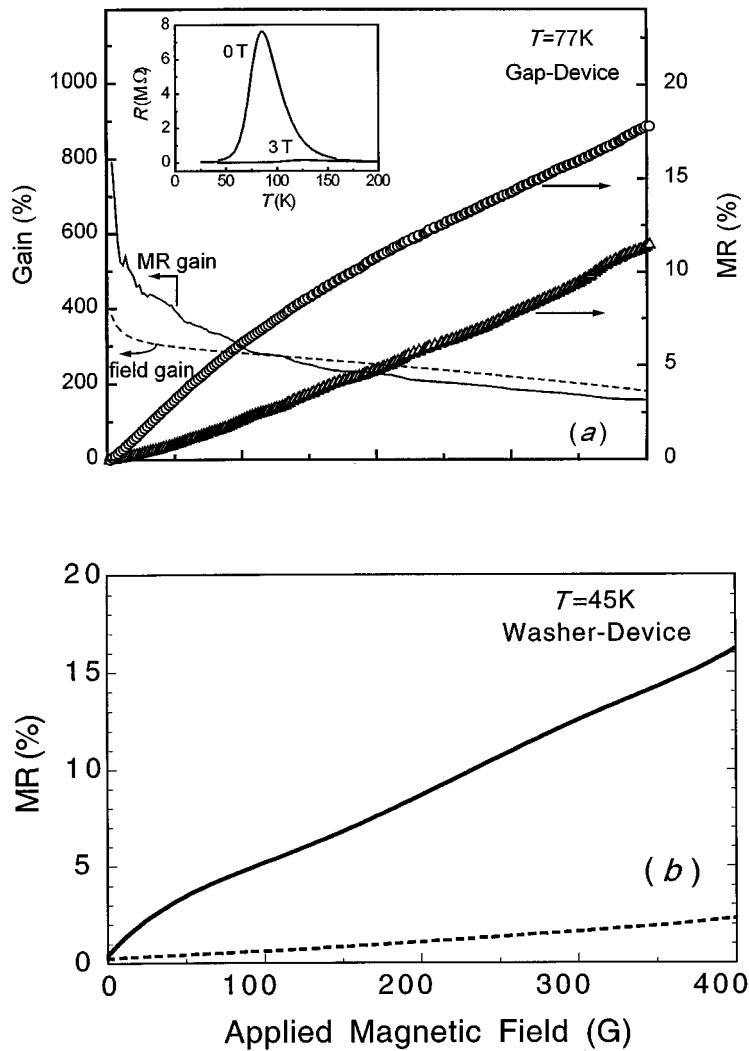


Figure 13. (a) Magnetic field dependence of MR effect with (open circles) and without (open triangles) YBCO for a gap-device at 77 K. The dimension of the NSMO bridge is $120\ \mu\text{m}$ by 1 mm. Both MR (solid line) and field (dashed line) gains are also plotted against the applied magnetic field. The inset shows typical R - T curves of an NSMO layer in a heterostructure of YBCO-NSMO without flux focusing at 0 and 3 T applied magnetic field. The MR is greater than 99.9% at 80 K. (b) Magnetic field dependence of MR effect with (solid line) and without (dashed line) YBCO for a washer-device at 45 K.

(d) *Low temperature HTS-CMR hybrid devices*

Since the properties of the CMR materials are quite spectacular at reduced temperatures, i.e. below 100 K, there may be some advantages to integrating them with HTS devices. In fact the SFQ logic at 4 K seems very attractive today with potential for this to make an impact even at 77 K. However, the real technology will be realized only when one can integrate SFQ logic circuits with semiconductor devices,

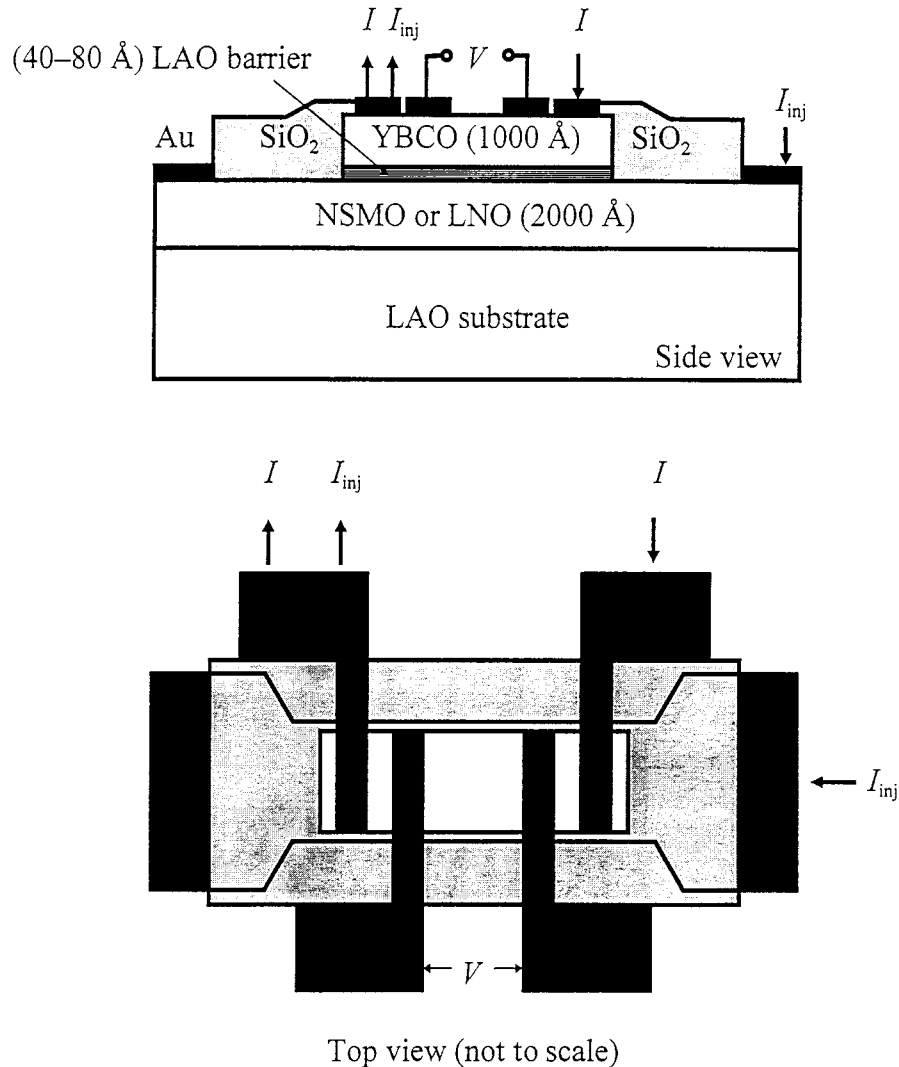


Figure 14. Schematic of a quasi-particle injection device and a circuit for the injection measurements. Vertical transport measurements for the characterization of the LAO barrier can also be performed.

and possibly cooled CMOS in order to overcome the large memory requirements of fast computation systems where the superconducting circuit will be primarily used for computation. The matching of the SFQ circuits which operate at millivolt levels and the CMOS at volt level presents a major problem and there is a real need for a transformer between the two systems. A CMR-based sensor which has large magnetoresistance at fields of the order of 0.2 T can provide a flux to voltage transformer at these temperatures (Kimura *et al.* 1996). A 1000 Å CMR ring detector will have at its centre a field of 0.2 T subtended by a vortex from the SFQ logic chip which can produce a large resistive modulation. Thus we can use the CMR as a flux to voltage converter.

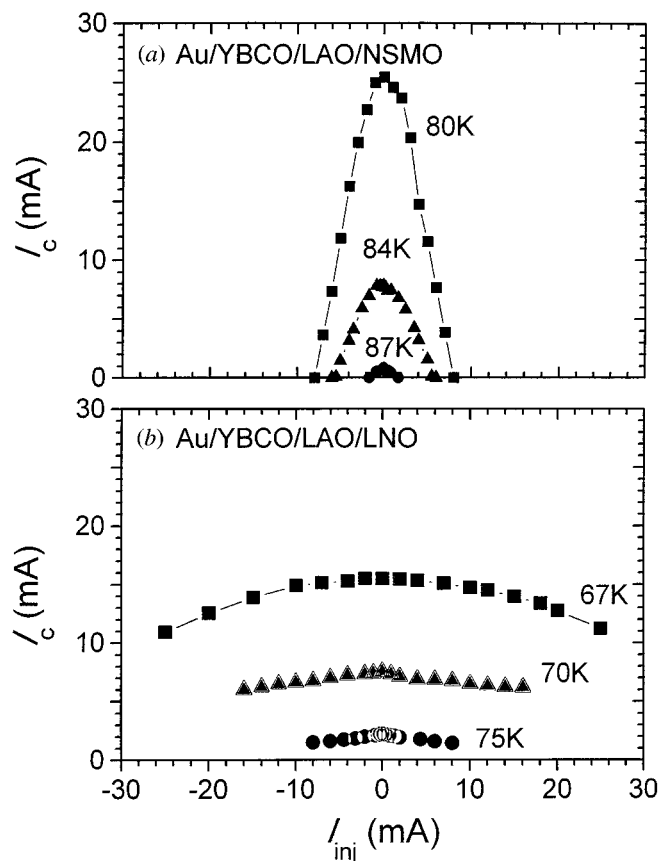


Figure 15. Dependence of the critical current of both Au–YBCO–LAO–NSMO (a) and Au–YBCO–LAO–LNO (b) QPIDs on the injection current at different temperatures. The current gain of 3.2, at 80 K, was obtained for Au–YBCO–LAO–NSMO (S–I–F) devices, which is about 10–30 times higher than that of Au–YBCO–LAO–LNO (S–I–N) devices.

Conversely, the Meissner effect of the superconductor can be used as a magnetic lens to enhance the flux coupling to a CMR detector thereby enhancing its sensitivity. In figure 12 is shown the schematic of an HTS magnetic lens coupled with a CMR resistor and the enhancement is obvious in figure 13 (Dong *et al.* 1996). Large gains of the order of 50 may be achievable with appropriate design of the device.

One of the most exciting new directions to emerge has been the area of spin injection into superconductors. Using CMR materials as ferromagnetic layers epitaxially grown with YBCO layers, FET type structures have been fabricated, where spin polarized electrons were injected from the manganite layer into the superconducting channel (Vas'ko *et al.* 1997; Dong *et al.* 1997). A typical device is shown in figure 14, and its I – V characteristics as a function of gate current is shown in figure 15 (Dong *et al.* 1997). The results can be directly compared to a gate where the electron source is unpolarized, where the CMR layer is replaced by a layer of metallic LaNiO_3 (LNO). In figure 15 is shown the effect of the spin polarized and unpolarized electrons on the critical current and clearly a 10–30-fold enhancement in pair breaking efficiency is

seen for the case of the polarized electron injection. These experiments take a larger meaning in the context of current theories of HTS where one is concerned about *s* versus *d* wave superconductors (Pines & Monthoux 1995), as well as the concept of spin charge separation in these superconductors (Si 1997). Similar experiments involving different superconducting channels are likely to throw more light on these issues. Thus spin injection in HTS may lead not only to potentially new devices but also may serve as a useful vehicle for understanding some of the forefront theoretical ideas.

3. Conclusion

In conclusion, the CMR materials are technologically exciting not just from the point of view of magnetoresistance but also because of their other peculiar properties. Conversely, many of these device approaches themselves may shed light on our understanding of the various mechanisms underlying these material properties.

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Discussion

R. STROUD (*Naval Research Laboratory, Washington, USA*). On the bolometers, were the noise measurements made on semiconducting substrates, or were those on lanthanum aluminate?

T. VENKATESAN. Either lanthanum aluminate or neodymium gallate—various types of substrates.

R. STROUD. Has Dr Venkatesan done any measurements made on semiconducting substrates yet?

T. VENKATESAN. No.

R. STROUD. Given that the devices are to be used uncooled, what hope is there of moving the peak in TCR, which is around 250 K, to a higher temperature?

T. VENKATESAN. If you take neodymium gallate and put LCMO on it, you can push it up to about 270 K or so. Actually it turns out that when you build a bolometer you do want to cool it a bit, because it's a transition edge device and you have to hold it at a given temperature. With a thermoelectric cooler you can cool it down to 250 K, which for these guys is room temperature.

R. STROUD. 270 K is room temperature. For every 20° you have to increase the number of thermoelectric coolers you have to put on it, so it gets much more expensive.

T. VENKATESAN. What I am saying is that I think these devices could be built fairly effectively around 270 K.

R. STROUD. On the spin injection devices, what happens if the lanthanum nickelate devices are measured at the higher temperatures that the NSMO devices are measured?

T. VENKATESAN. The problem is the following: we have a lot of experience with the YBCO/NSMO type of stuff, so we can make very high quality YBCO. With lanthanum nickelate, the quality is a little bit reduced and so measurements were done at lower temperature.

R. STROUD. How does the number of quasi-particles that are injected in each case compare? What is the yield of quasi-particles?

T. VENKATESAN. They are identical devices and identical geometries so the number to really compare is the current ratio, which is a good way to look at it. We have calculated it, but I don't have that number here and I don't want to guess.

M. BLAMIRE (*University of Cambridge, UK*). The current gain is an important parameter, but at what voltage are the quasi-particles being injected, because the power gain is an important issue? Can this be discriminated from heating?

T. VENKATESAN. We have done an experiment in the last week where we varied the barrier layer (which is lanthanum aluminate) and what we find is that the gain is like a bell-shaped function. In other words, the gain for very thin barriers is very low and as we make it thicker it goes to a maximum and then it comes back down if you make the barrier even thicker. That seems to rule out thermal effects; if you believe in thermal effects then the largest barrier should give the maximum effect and that's not what we see.

T. T. M. PALSTRA (*University of Groningen, The Netherlands*). In the other papers in this volume, there has been much attention paid to the spin disorder near the surface of these materials, but in the materials mentioned by Dr Venkatesan, there is full spin polarization with photoemission which probes on a depth scale of only about 5 Å. How does he explain that? Is there is no spin disorder at the surface?

T. VENKATESAN. If you take a SQUID to measure the entire film, and then look within 5 Å of the surface, what you progressively see is that the magnetization is deteriorating. The temperature dependence of the magnetization is changing: at low temperatures they all look the same; as the temperature is increased, it falls off much more as you look at the surface, indicating that there is some kind of degradation in the magnetic behaviour of the material at the surface.

T. T. M. PALSTRA. But would he argue that there is no spin disorder at low temperatures?

T. VENKATESAN. There may be other explanations for this—why have a depth-dependent effect? I'm not making up my mind about it right now.

A. J. MILLIS (*The Johns Hopkins University, USA*). The question he is asking is that at zero temperature the photoemission experiments show that the surface is nice, which appears to contradict data presented elsewhere.

T. VENKATESAN. The spin polarization of the surface is definitely 100%, which is true at low temperatures, up to 40 K in LSMO, and then it drops off.

A. MAIGNAN (*Laboratoire CRISMAT, Université de Caen, France*). It is true that the photoemission measures just 5 Å and there is 100% spin polarization at low temperature, but the density of states for the majority band is very small and that might be due to spin disorder.

A. J. MILLIS. Why is there a difference between *c*-axis YBCO and *a*-axis YBCO in this injection business?

T. VENKATESAN. These are all recent data—we don't understand the difference. I have just put up all the data to show that there is a lot of interesting physics here and the spin polarized tunnelling may be a way to really probe it.

A. J. MILLIS. May I ask Dr Venkatesan to clarify the experiment a little bit? The difference between *a*-axis and *c*-axis, in which direction is the injected current going?

T. VENKATESAN. The injected current comes down the *a*-axis and the channel current is flowing along the *b*-axis direction in the plane. In the *a*-axis device the *c*-axis

lies in the plane of the film so you can define your device along the b -axis or along the c -axis; the work that we did was along the b -axis, we just wanted to see whether that replicated the other one. Much to my surprise it was completely different, so now we want to go and do the other direction.

M. BLAMIRE. Dr Venkatesan's plot of his deduced surface magnetization is exactly of the form in Mathur *et al.*'s (1997) paper deduced from our experiments. In other words the linear slope of magnetization against temperature is what we would also require to get fits to the grain boundary devices.

T. VENKATESAN. One of the comments made was that in the mesoscopic model it is difficult to explain the temperature dependence of the tunnelling. In other words a tunnelling model would have a hard time to explain the temperature dependence. But a tunnelling model makes an assumption that the polarization is 100% at all temperatures below T_C , and what we see here is that the polarization is actually decaying with temperature; I would be very curious to take some of the device data and plot them on top of this. I have a feeling that these data pretty much explain the temperature dependence.

M. BLAMIRE. I would agree, but it is true also that your information on the spin polarization comes from the very thin surface layer. So if you are thinking about spin polarized tunnelling, you should be concerned with the spin polarization within the bulk of the crystal which can then tunnel through this thin layer. So they are not actually the same thing.

T. VENKATESAN. I agree. If you build a trilayer device, how much does the 5 or 10 Å layer control the transport properties? That's really the issue, and I don't have the answer to that, but I think what this result says is that we should take this into account.

Additional references

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