Intrinsic Tunneling in Cuprates and Manganites

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The most anisotropic high temperature superconductors like Bi₂Sr₂CaCu₂O₈, as well as the recently discovered layered manganite La₁₄Sr₁₆Mn₂O₇ are layered metallic systems where the interlayer current transport occurs via sequential tunneling of charge carriers. As a consequence, in Bi₂Sr₂CaCu₂O₈ adjacent CuO₂ double layers form an intrinsic Josephson tunnel junction while in in La₁₄Sr₁₆Mn₂O₇ tunneling of spin polarized charge carriers between adjacent MnO₂ layers leads to an intrinsic spin valve effect. We present and discuss interlayer transport experiments for both systems. To perform the experiments small sized mesa structures were patterned on top of single crystals of the above materials defining stacks of a small number of intrinsic Josephson junctions and intrinsic spin valves, respectively.

74.50, 85.25, 75.45, 75.70

I. INTRODUCTION

It is well established that the most anisotropic high temperature superconductors form superconducting multilayers where the superconducting unit (single layers, bilayers or trilayers of CuO₂) are separated by insulating barrier layers. Examples are Bi₂Sr₂CaCu₂O₈ (BSCCO) or Tl₂Ba₂Ca₂Cu₃O₁₀. Interlayer transport occurs via sequential tunneling of charge carriers, and consequently suitably patterned structures can be considered as a stack of intrinsic Josephson tunnel junctions (cf. Fig. 1). The intrinsic Josephson effect has been intensively investigated within the last decade for a number of reasons. For example, the gap voltage limiting the high frequency properties of intrinsic Josephson junctions is in the range of 30 mV allowing ac Josephson currents at frequencies of several THz. Thus, such junctions are highly promising for THz applications like oscillators or mixers. Second, a number of fundamental phenomena can be investigated like nonequilibrium effects due to charge imbalance in the superconducting layers or collective effects like Josephson plasma oscillations involving the stack as a whole, the collective dynamics of Josephson fluxons, or the formation of charge solitons. A third issue is to use intrinsic Josephson junctions for tunneling spectroscopy. Their advantage over artificial tunnel junctions is to probe not only the surface layer but all layers within the stack. The prize, however, is that always a number of layers are measured in series. Thus the information provided is essentially the average over several layers. Using this method phonons in Bi- and Tl-based cuprates have been detected. Recently, the method has been used to investigate the quasiparticle density of states in both the superconducting and normal regime.

A system very similar to the high temperature superconductors are the layered manganites like La₃Sr₂Mn₂O₇. Here, MnO₂ bilayers are separated by thin layers consisting of La and Sr ions. In La₃Sr₁₆Mn₂O₇ (LSMO) the MnO₂ bilayers undergo a metal-insulator transition near 90 K while the intervening layers remain insulating. Within each MnO₂ sheet magnetic moments are ferromagnetically ordered. As revealed by neutron diffraction measurements, adjacent sheets become antiferromagnetically arranged below 70 K with magnetization vector perpendicular to the layers. Interlayer transport of the spin-polarized charge carriers occurs via tunneling processes, leading to a low temperature tunneling magnetoresistance (TMR) in addition to the effect of Colossal Magnetoresistance (CMR) observed near the metal-insulator transition (cf. Fig. 2). Thus, at low temperatures, adjacent MnO₂ bilayers can be considered as a natural TMR element or spin valve switching from large to small tunneling resistance upon
application of an external magnetic field. Consequently, a suitably patterned single crystal acts as an intrinsic stack of spin valves [18].

FIG. 1. High temperature superconductor Bi$_2$Sr$_2$CaCu$_2$O$_8$ intrinsically forming a stack of Josephson junctions (a) and layered manganite La$_{1.4}$Sr$_{1.6}$Mn$_2$O$_7$ intrinsically forming a stack of spin valves (b). S denote superconducting, FM ferromagnetic and I insulating layers. Arrows in (b) denote direction of magnetization at low temperatures.

What is a suitable size of an intrinsic Josephson junction stack or an intrinsic spin valve stack to be investigated? In terms of intrinsic Josephson junctions in BSCCO there are two limitations. First, the Josephson length $\lambda_J$ determining, e.g., the size of a Josephson fluxon is in the order of 0.2 - 0.5 $\mu$m [1,2,8,9]. If the formation of such fluxons is to be avoided all lateral dimensions of the stack should not exceed $\lambda_J$. In case of a "long" Josephson stack still the smaller side of the stack should be below $\lambda_J$. The second requirement is that the stack should not contain too many junctions. For many fundamental investigations stacks of well below some tens of junctions are desirable in order to produce interpretable results. Also, for stacks containing a large number of junctions ohmic heating becomes severe. The thickness of the stack thus should be below some 10 nm.

In terms of the layered manganites hall probe measurements [19] and magneto-optical investigations [20] have revealed that LSMO single crystals exhibit a large number of magnetic domains typically some 10 $\mu$m in size. In order to observe clear spin valve effects, structures smaller than the size of such domains are desirable. For the same reasons as for intrinsic Josephson junctions also here the stacks should consist of only a few unit cells.

### II. SAMPLES AND MEASUREMENTS

BSCCO single crystals were grown from a stoichiometric mixture of the oxides and carbonates [21]. Epoxy was used to glue approximately $1 \times 1 \times 0.1$ $\mu$m$^3$ large crystals to a sapphire substrate. To obtain a sufficiently small contact resistance the crystals were cleaved immediately before mounting them into the vacuum chamber and the crystal surface was covered with silver. The contact resistance was typically $10^{-5}$ $\Omega$cm$^2$. Subsequently, rectangular mesa structures with minimal lateral dimensions of 0.5 $\mu$m were patterned using electron beam lithography and argon-ion milling. For electrical insulation of the lead contacting the top of the mesa a 250 nm thick SiO layer was evaporated. The top contact was provided by a 300-400 nm thick gold or silver layer. Currents were extracted from the base crystal using large pads contacting its top surface. The leads and contact pads were patterned by photolithography and argon-ion milling. The LSMO single crystals of roughly 1 mm$^3$ in size were grown using a floating zone technique [15,22]. Mesa structures were patterned using the same techniques as for the BSCCO single crystals. The only difference was that the crystal surface was initially polished mechanically instead of being cleaved. The mesa size ranged from $5 \times 5$ to $10 \times 10$ $\mu$m$^2$. Their thickness was about 20 nm corresponding to a stack of 20 spin valves. For both systems transport measurements were performed in a two-terminal configuration. Low pass filters were used to reduce external noise and the bias current was provided by a battery powered current source.

### III. RESULTS

#### A. Intrinsic Josephson junctions in Bi$_2$Sr$_2$CaCu$_2$O$_8$

In this section we will discuss some results obtained for intrinsic Josephson junction stacks. The main focus will be on quasiparticle tunneling in both the superconducting and the normal state.

FIG. 2. Current voltage characteristic at 4.2K of $1 \times 1 \mu$m$^2$ BSCCO mesa structure SH149 containing 5 junctions. Lower inset shows same characteristic on expanded scale, upper inset shows derivative $dI/dU$. 

\[ I(\mu A) \quad U(V) \]

\[ 0 \quad 0.5 \quad -50 \quad -0.1 \quad 0.1 \quad 1 \quad 0 \quad 0.1 \quad 0.5 \quad 1 \quad 0 \quad 0.1 \quad 0.5 \quad 1 \quad 0 \quad 0.1 \quad 0.5 \quad 1 \quad 0 \quad 0.1 \quad 0.5 \quad 1 \]

\[ I(mA) \quad U(V) \]

\[ 0 \quad 0.5 \quad -50 \quad -0.1 \quad 0.1 \quad 0.3 \quad 0.6 \quad 0.9 \quad -0.9 \quad -0.6 \quad -0.3 \]

\[ dI/dU(mS) \quad U(V) \]

\[ 0 \quad 0.5 \quad -50 \quad -0.1 \quad 0.1 \quad 0.3 \quad 0.6 \quad 0.9 \quad -0.9 \quad -0.6 \quad -0.3 \]

\[ dI/dU(mS) \quad U(V) \]

\[ 0 \quad 0.5 \quad -50 \quad -0.1 \quad 0.1 \quad 0.3 \quad 0.6 \quad 0.9 \quad -0.9 \quad -0.6 \quad -0.3 \]

\[ dI/dU(mS) \quad U(V) \]

\[ 0 \quad 0.5 \quad -50 \quad -0.1 \quad 0.1 \quad 0.3 \quad 0.6 \quad 0.9 \quad -0.9 \quad -0.6 \quad -0.3 \]
Fig. 3 shows a typical current voltage ($I-U$) characteristic of $1 \times 1 \mu m^2$ large mesa SH149 patterned on a slightly overdoped BSCCO single crystal with $T_c = 86$ K, as determined from the onset of interplane superconductivity. The mesa consisted of $N = 5$ junctions. Below the critical currents of these junctions the $I-U$ characteristic exhibits 5 branches in the resistive state differing by the number of resistive junctions (lower inset of Fig. 3). On the large current scale all junctions are resistive. The upper inset in Fig. 3 shows the conductance $dI/dU$. A clear gap structure is visible with a total gap value $2N\Delta = 0.25$ V (the sum of the gap voltages of all junctions) corresponding to $\Delta = 25$ meV. Below the gap the conductance is U-shaped. Also note the dip and hump feature above 2$\Delta$ typical for BSCCO tunneling spectra [23]. In Fig. 3 we show conductance curves of the same mesa at various temperatures. While the amplitude of the gap peak strongly decreases and the dip and hump features disappear when approaching $T_c$, the voltage position of the gap changes only weakly and almost continuously transits into the pseudogap regime.

![Fig. 3. Conductance $dI/dU$ of BSCCO mesa SH149 at temperatures between 4.2 K and 140 K. From 10 K to 140 K temperature is increased in steps of 10 K. In addition curve at $T_c$ is shown. Curves are vertically offset in steps (140 K - $T$)/20.](image)

This can be seen more clearly in Fig. 4 where we plot $\Delta$ vs. $T$ for mesa SH149 as well as for 6 junction $0.8 \times 0.8 \mu m^2$ mesa SH146. Also this mesa was slightly overdoped with a $T_c$ of 81 K. For both mesas $\Delta(T)$ exhibits a minimum near $T_c$. While in the superconducting state $\Delta$ is almost identical for both mesas they differ strongly in both magnitude and temperature dependence of $\Delta$ above $T_c$. When applying a magnetic field perpendicular to the layers we found that $\Delta$ decreases in the superconducting state while it is almost field independent or even increases slightly with field in the pseudogap regime.

![Fig. 4. Peak voltage (divided by 2N) of conductance curves of conductance curves vs. $T$ for the two slightly overdoped BSCCO mesas SH149 ($T_c = 86$ K) and SH146 ($T_c = 81$ K). Lines correspond to BCS temperature dependence of the gap.](image)

An important question is to what extent the two phenomena gap and pseudogap are related. To our opinion, the intrinsic tunneling data suggest they are not for several reasons. In contrast to the gap in the superconducting state the pseudogap is strongly material dependent. The dip in $\Delta(T)$ near $T_c$ even gives the feeling that two phenomena compete with each other. In terms of magnetic field dependence gap and pseudogap clearly behave differently. We also investigated slightly under-
doped samples where we found that $\Delta$ almost goes to zero at $T_c$. For these samples gap and pseudogap were observable simultaneously in the superconducting state. Although the above features - similar observations have also been made by other groups investigating intrinsic tunneling in BSCCO - are certainly not a proof of the independence or even competition of two unrelated phenomena, they at least raise doubts of a common mechanism leading to the superconducting gap and to the pseudogap.

**B. Intrinsic spin valves in La$_{1.4}$Sr$_{1.6}$Mn$_2$O$_7$**

Next we turn to the properties of mesa structures patterned on LSMO single crystals. In contrast to (transport) measurements on bulk single crystals the mesa technique provides the possibility to probe a small region well below the size of a magnetic domain. We will show that such mesas indeed exhibit the properties expected for a stack of intrinsic spin valves.

![Fig. 6. Current voltage characteristics of 5 $\times$ 5 $\mu$m$^2$ large mesa DDA4/4 patterned on a LSMO single crystal in magnetic fields between 0 and 0.7 T. The field, oriented perpendicular to the layers, is increased in steps of 0.1 T. Inset: derivative $dI/dU$ for $B = 0$ (solid line) and for $B = 0.7$ T (dashed line).](image)

Fig. 6 shows field dependent 4. K $I$-$U$ characteristics of the 5 $\times$ 5 $\mu$m$^2$ large mesa DDA4/4 having a thickness of about 20 nm. The resistance is clearly lowered with increasing magnetic field. The inset shows the conductance $dI/dU$ for $B = 0$ and $B = 0.7$ T. There is a conductance maximum at 1.4 V for $B = 0$, corresponding to roughly 75 mV per spin valve (with a 50% error margin, since we do not know the number of layers in the mesa precisely) slightly shifting to 1.3 V for $B = 0.7$ T. The overall conductance curves look strikingly similar to the low temperature tunneling spectra of BSCCO mesas (cf. Fig. 2) although the similarities might be accidental. A possible reason for the dip structure in the LSMO mesa might be the excitation of spin waves. We also note that similar conductance curves as in Fig. 2 are obtained for temperatures up to the metal-insulator transition, with a slight ($\sim 10 - 20\%$) decrease in the conductance maximum.

What can be expected for the low temperature magnetoresistance of a mesa probing a single magnetic domain? In zero field, the magnetization vectors of adjacent MnO$_2$ bilayers are aligned antiparallel (cf. Fig. 2). In magnetic fields parallel to the layers the magnetization will tilt continuously towards parallel orientation leading to a continuous decrease of tunneling resistance. In perpendicular fields there should be a spin flop from antiparallel to parallel for a field where the Zeeman energy ($\propto HM$) overcomes the interlayer coupling energy ($\propto M^2$). Consequently, there should be a discontinuous jump in magnetoresistance for perpendicular fields.

![Fig. 7. Magnetoresistance of LMSO mesa QU10/6 for field orientation parallel (solid symbols) and perpendicular (open symbols) to the layers. Bias current is set in the subgap regime of the current voltage characteristic. Inset illustrates magnetization vectors for zero field, parallel field and perpendicular field above the threshold field where the jump in $R(B)$ occurs.](image)

Fig. 7 shows data for the 10 $\times$ 10 $\mu$m$^2$ large mesa QU10/6 for both field orientations. In parallel fields $R(B)$ indeed decreases continuously while in perpendicular fields one jump near 0.3 T is observed with a small hysteresis of about 30 mT for increasing/decreasing fields. We observed similar data for in total 10 mesas. In perpendicular fields the jump in magnetoresistance was observed in fields between 50 mT and 0.4 T, without apparent correlation to mesa size or thickness. The observed range of switching fields corresponds nicely to magneto-optical measurements on LSMO single crystals where first order spin flop transitions have been observed between 0.11 and 0.48 T. From the fact that only one single switching event is visible in $R$ for perpendicular fields we conclude that all spin valves within the stack switch collectively (i.e. the magnetization vectors of all MnO$_2$ bilayers flip simultaneously). Note, for comparison, that intrinsic Josephson junctions in BSCCO can be switched to the resistive state one by one. For a more detailed discussion of the magnetoresistance observed for our mesa structures, see [13].
IV. CONCLUSIONS

The above data may have shown that there are a number of similarities between the cuprate Bi$_2$Sr$_2$CaCu$_2$O$_8$ (BSCCO) and the layered manganite La$_{1.4}$Sr$_{1.6}$Mn$_2$O$_7$ (LSMO). In both systems interplane transport occurs via sequential tunneling of charge carriers. Suitably prepared mesa structures on BSCCO and LSMO single crystals act as stacks of intrinsic Josephson tunnel junctions and spin valves, respectively. Both systems allow the investigation of a number of electrodynamic effects like Josephson fluxon dynamics or the collective switching of spin valves.

From a microscopic point of view there is a striking similarity between the tunneling spectra of both systems at low temperatures. Further investigations will show to what extent a comparison between the two materials will lead to an improved understanding of both the high temperature superconductors and the manganites.

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