Magnetoresistive Effects in Ferromagnet-Superconductor Multilayers

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We consider a nanoscale system consisting of Manganite-ferromagnet and Cuprate-superconductor multilayers in a spin valve configuration. The magnetization of the bottom Manganite-ferromagnet is pinned by a Manganite-antiferromagnet. The magnetization of the top Manganite-ferromagnet is coupled to the bottom one via indirect exchange through the superconducting layers. We study the behavior of the critical temperature and the magnetoresistance as a function of an externally applied parallel magnetic field, when the number of Cuprate-superconductor layers are changed. There are two typical behaviors in the case of a few monolayers of the Cuprates: a) For small magnetic fields, the critical temperature and the magnetoresistance change abruptly when the flipping field of the top Manganite-ferromagnet is reached. b) For large magnetic fields, the multilayered system re-enters the zero-resistance (superconducting) state after having become resistive (normal).

Magnetoresistive effects in ferromagnet/normal-metal multilayers have been studied since the early 90’s, where oscillations in exchange coupling and magnetoresistance have been observed [1]. In this manuscript, we are interested in magnetoresistive effects in the spin-valve configuration of ferromagnet/superconductor multilayers. A spin-valve system consists of a pair of ferromagnetic layers anti-ferromagnetically coupled and separated by a metal spacer where one layer is “pinned,” or fixed in magnetization by an adjacent anti-ferromagnetic layer. Since electrons moving in the metal spacer encounter less resistance if the magnetizations of the layers are aligned, a spin-valve system can exhibit a magnetoresistance effect when an external magnetic field is applied that is strong enough to flip the unpinned layer. If ordinary low critical temperature \( T_c \) superconducting metals are used as spacers of thickness less than a 130Å, the proximity effect due to many lattice matched ferromagnets will be so strong as to destroy superconductivity, and thus any magnetoresistive effect associated with a superconducting metal spacer will be absent [2]. If one insists on using a low \( T_c \) superconducting metal it is necessary to increase its thickness more than 130Å and use a weaker lattice matched ferromagnet. This situation was investigated experimentally in CuNi/Nb multilayers, where a very weak magnetoresistive effect was found [3]. The ferromagnetic proximity effect is reasonably small such that it preserves superconductivity, but the magnetoresistive effect is also small as the Nb spacer involves many monolayers [3]. However, if the spacer layers are of thickness less than 130Å and consist of metal Copper oxides like the high-\( T_c \) d-wave superconductors and the ferromagnets consist of Manganese oxides (CMR-materials), then the ferromagnets can antiferromagnetically couple through the superconductor given that the proximity effect due to ferromagnets is not strong enough to suppress the superconducting state [4]. Since it has been experimentally demonstrated that a single monolayer of a Copper oxide can be superconducting with a high \( T_c \) [5], and that magnetic coupling between CMR-ferromagnets separated by a complex oxide is possible [6], we propose in this manuscript that the critical temperature and the resistance for a CMR-ferromagnet/high-\( T_c \)-superconductor multilayer in such a spin-valve configuration can change dramatically as a function of applied magnetic field.

Previous theoretical works that studied proximity and magnetoresistive effects in F/S/F multilayers have focused on standard ferromagnets and standard superconductors and have used continuum Usadel equations [7, 8]. Unfortunately, magnetoresistive effects in these systems have been demonstrated experimentally to be small [8]. By contrast, we proceed by analyzing the dependence of the superconducting critical temperature \( T_c \) of such Manganese-oxide-ferromagnet-Copper-oxide-superconductor spin-valve systems as a function of externally applied magnetic field. First, we assume that the Curie temperature of the ferromagnets \( T_F \gg T_c \), where \( T_c \) is the critical temperature of the superconductor spacer. Typical values are \( T_F \approx 300K \) and \( T_c \approx 85K \). This condition implies that the magnetization of each ferromagnet is essentially saturated when \( T \approx T_c \), and thus that the magnetic part of the free energy is essentially independent of temperature for \( T \approx T_c \). We use a Ginzburg-Landau model (which can be derived rigorously for a d-wave superconductor) to write the free energy density of each of the \( n \) superconducting layers in terms of the superconducting order parameter \( \Psi(\mathbf{r}) \). The free energy for the \( j \)th superconducting layer is

\[
\mathcal{F}_{sj} = \mathcal{F}_{o_j} + \int dA \left[ \alpha_j |\Psi_j|^2 + \frac{\beta_j}{2} |\Psi_j|^4 + \gamma_j \nabla \Psi_j |^2 \right],
\]

where \( \beta_j > 0, \alpha_j = \alpha_{o_j}(T - T_c^{(o)}) \) and \( T_c^{(o)} \) is the critical temperature of the superconducting material comprising an isolated layer \( j \).

Since we will also be considering the effect of a uniform magnetic field applied parallel to the plane of each layer, we add a term \( \delta_j |\mathbf{B}_{tot}|^2 |\Psi_j|^2 \) to each superconductor’s free energy. This term accounts for the coupling with the spin degrees of freedom of the superconductor. Note
that $\delta_j > 0$ since a magnetic field introduces an energy cost related to the superconducting electrons, which are paired in singlet states. Also note that the coupling of the parallel magnetic field with the charge degrees of freedom is not relevant for small magnetic fields as the thickness of the superconducting spacer is less than 130 Å. We assume that the ferromagnets surrounding the superconducting spacer are identical, producing an exchange field of magnitude $B_F$ felt only by neighboring superconducting layers (the ferromagnetic proximity effect), and that the external field is applied such that it is aligned with the magnetization of the pinned layer. For the layer closest to pinned ferromagnet (see fig. 1), then $B_{tot\,1} = B_{ext} - B_F$ while all interior layers have simply $B_{tot\,j} = B_{ext}$ where $j = 2 \ldots (n - 1)$. Most importantly, though, if $B_{flip}$ is the critical applied field value that flips the magnetization of the unpinned layer, the field felt by the superconducting layer adjacent to the unpinned ferromagnet is

$$B_{tot\,n} = \begin{cases} B_{ext} + B_F & B_{ext} < B_{flip} \\ B_{ext} - B_F & B_{ext} > B_{flip} \end{cases}$$

For the systems we will analyze, $B_{flip} \ll B_F$ which is crucial in that a small applied field may greatly affect the resistance of the system. Since we are considering parallel magnetic fields (with no components perpendicular to the superconducting layers), we will take each superconducting order parameter $\Psi_j$ to be spatially uniform, making the terms in (11) which depend on the gradient of $\Psi_j$ zero.

To account for the Josephson coupling between superconducting layers, we add terms of the form $c_{ij} |\Psi_i - \Psi_j|^2$ to the free energy, but only consider coupling between adjacent layers. Furthermore, we only consider the case where the $c_{ij}$’s are nonnegative, i.e. there is no $\pi$-coupling as found when superconducting layers are separated a ferromagnetic layer.

To calculate the superconducting critical temperature of the system, taking all superconducting layers to be identical, we first write $\tilde{\delta} = \delta/\alpha_o$ and $\tilde{\epsilon} = \epsilon/\alpha_o$, and then seek the temperature for which $F_s$ is no longer a local minimum at $\Psi = 0$. For $\Psi$ near zero, the free energy varies by an amount proportional to

$$\sum_{ij} \alpha_o \Psi_i^* F_{ij} \Psi_j + \sum_j \beta_j |\Psi_j|^4 / 2$$

where the nonzero elements of the tridiagonal matrix $\tilde{F}$ are given by

$$F_{ij} = \begin{cases} T - T_{c(0)} + \tilde{\delta}|B_{tot}|^2 & i = j \\ +\tilde{\epsilon}_{i-1} + \tilde{\epsilon}_{i+1} & |i - j| = 1 \\ -\tilde{\epsilon}_{ij} & \text{else} \end{cases}$$

The system spontaneously enters the superconducting state only when $\tilde{F}$ has negative eigenvalues, and so we may calculate the superconducting critical temperature of the system by finding the largest value of $T$ that makes $\det(\tilde{F}) = 0$.

First consider only a single monolayer of a Copper oxide superconductor between CMR-ferromagnets in the spin-valve configuration. The critical temperature becomes, simply

$$T_c = T_{c(0)} - \tilde{\delta}|B_{tot}|^2$$

The magnetic proximity effect (exchange field present) in the superconducting layer has its origins in the superexchange effect between Manganese and Copper ions through common oxygen atoms shared in the perovskite structure of the multilayer. The superexchange mechanism produces an antiferromagnetic coupling between the Manganese and Copper spins, and thus the exchange field produced in the superconducting layer by each ferromagnet is always anti-aligned with the magnetization.
temperatures within a reasonably large range (10% of normal for fields slightly above temperature within this range below $T_c$). For a large enough applied field ($B_{\text{ext}} > B_{\text{flip}}$), the system re-enters the superconducting state by several orders of magnitude from zero ($B_{\text{ext}} < B_{\text{flip}}$), while for the one-layer case the change is very much larger ($2B_{F}$).

Even at zero field, the critical temperature of the two-layer system is less than $T_c(0)$ because a non-zero exchange field is already present in each superconducting layer. Then, once $B_{\text{ext}}$ exceeds $B_{\text{flip}}$, the change in $T_c$ is much smaller than in the one-layer case because the magnitude of the field felt by the superconductor nearest to the flipping ferromagnet changes only by $B_{\text{flip}}$ in the two-layer case, while for the one-layer case the change is very much larger ($2B_{F}$).

Finally, two-layer systems whose temperature are in the window of opportunity ($T_0 < T < T_{\text{flip}}$) are normal first and then become superconducting for $B_{\text{ext}} > B_{\text{flip}}$, after which they remain superconducting until the external field becomes extremely large ($> B_{F}$). Thus the superconducting-normal-superconducting re-entrant behavior seen at relatively low fields for properly cooled one-layer systems is basically no longer present in the two-layer system (except for temperatures in a very small range below $T_0$).

For systems having more than two superconducting monolayers, the behavior of $T_c$ is qualitatively very similar that of the two-layer system. However, as the number of superconducting layers is increased, the sign of magnetic coupling between ferromagnetic layers starts to oscillate between ferromagnetic and antiferromagnetic [4]. In addition, in the anti-aligned cases, $B_{\text{flip}}$ is reduced due to the weakened anti-ferromagnetic coupling between the ferromagnetic layers. As the number of superconducting layers gets larger there is essentially no difference in $T_c$ for parallel on anti-parallel orientations of the ferromagnets. Furthermore, $T_c$ starts to reach its three-dimensional bulk value $T_c^{(3D)}$ as the effects of the ferromagnets become confined to the surfaces of the superconductor, and thus become relatively speaking much weaker than in the case of a few monolayers.

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\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{phase_diagram.png}
\caption{Schematic (a) phase diagram for two YBCO superconducting layers between LCMO ferromagnets and (b) the resistivity for current applied parallel to the layers. For this particular system, $T_c = 82.90$ K for $B_{\text{ext}}$ slightly less that $B_{\text{flip}}$ and $T_{\text{flip}} = 83.01$ K. The $\delta$ and $B_F$ parameters are the same as those used in fig. 2 but also $\tilde{\epsilon}_{12}$ has been taken to be about 15% of $T_c(0)$, or 12.75 K.}
\end{figure}

of the respective ferromagnet (see fig. 1). Now, while the ferromagnetic layers surrounding the superconductor are still anti-aligned in magnetization ($B_{\text{ext}} < B_{\text{flip}}$), the superconductor is unaffected by the ferromagnets, whose exchange fields cancel each other. However, for $B_{\text{ext}} > B_{\text{flip}}$, the superconductor feels the combined magnetic field of the aligned ferromagnets. For the single layer case, then

$$B_{\text{tot}} = \begin{cases} B_{\text{ext}} & B_{\text{ext}} < B_{\text{flip}}, \\ B_{\text{ext}} - 2B_{F} & B_{\text{ext}} > B_{\text{flip}}, \end{cases}$$

such that the phase diagram for the system behaves as in fig. 2 showing a dramatic change in the critical temperature at $B_{\text{ext}} = B_{\text{flip}}$. This abrupt change in $T_c$ provides for a super-colossal magneto-resistive effect, where the magneto-resistance of the spin-valve system can change by several orders of magnitude from zero ($B_{\text{ext}} < B_{\text{flip}}$) to metallic values ($B_{\text{ext}} > B_{\text{flip}}$) for systems cooled to temperatures within a reasonably large range (10% of $T_c$ or larger) (see fig. 2). Though a system cooled to a temperature within this window of opportunity becomes normal for fields slightly above $B_{\text{flip}}$, our model also predicts that the system re-enters the superconducting state for a large enough applied field ($B_{\text{ext}}$ anywhere from $B_{\text{flip}}$ to $2B_{F}$, depending on the temperature of the system).

For two superconducting monolayers deposited between anti-aligned ferromagnetic layers, the critical temperature is

$$T_c = (T_1 + T_2)/2 + \sqrt{(T_1 - T_2)/2}^2 + \tilde{\epsilon}_{12}^2.$$  

where $T_{1,2} \equiv T_c(0) - \delta [B_{\text{tot}}{1,2}]^2 - \tilde{\epsilon}_{12}$.

The phase diagram of a representative two-layer system is shown in fig. 3. While the two-layer system exhibits effects like those seen for the one-layer, including the abrupt change in $T_c$ around $B_{\text{flip}}$, there are some key differences between the systems.

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