PHYSICAL REVIEW B **67**, 020411(R) (2003)

Transport spin polarization in $SrRuO_3$ measured through point-contact Andreev reflection

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(Received 2 August 2002; published 31 January 2003)

We report a study in which Andreev reflection using a Nb point contact is used to measure the transport spin polarization of the $4d$ itinerant ferromagnet $SrRuO_3$ in the ballistic limit of transport. The degree of transport spin polarization is comparable to that of the hole-doped rare-earth manganites. We conclude that the large transport spin polarization results mainly from a difference in the Fermi velocities between the majority- and minority-spin channels in this material.

DOI: 10.1103/PhysRevB.67.020411 PACS number(s): 75.70.−i, 72.25.−b, 73.40.−c

Metallic oxide ferromagnets have been a field of great interest since the discovery of colossal magnetoresistance in doped rare-earth manganites.[1,2] Though the initial interest was triggered by the phenomenon of colossal magnetoresistance, it was soon realized that the large degree of spin polarization observed in many of these oxides also made them potential candidates to explore novel forms of electronics where both the charge and spin of the electrons could be used.[3,4,5] Towards this end, ferromagnetic tunnel junctions exhibiting both large positive and negative magnetoresistance using ferromagnetic oxides have already been fabricated.[3] Another field of great interest has been the effect of spin injection on the critical current of superconductors. In this class of experiments, spin-polarized carriers from an oxide ferromagnet are typically injected into a *d*-wave superconductor such as $YBa_2Cu_3O_7$ while the suppression in critical current of the superconductor is measured. While the ability to grow epitaxial superconducting layers on many of the perovskite oxide ferromagnets makes them attractive sources of spin-polarized electrons for these experiments, quantitative interpretation of the data is crucially dependent on prior knowledge of the degree of spin polarization of the injected carriers across the ferromagnet/superconductor interface. This can, however, be difficult to calculate, since most perovskite oxide ferromagnets have complicated band structures with several bands crossing the Fermi surface, and further Fermi surface fragmentation often results from subtle crystalline distortions. It is therefore important to measure the degree of spin polarization in these materials experimentally.

The spin polarization in a ferromagnet is normally defined as $P=[N_\uparrow(E_F)-N_\downarrow(E_F)]/[N_\uparrow(E_F)+N_\downarrow(E_F)]$, where $N_\uparrow(E_F)$ $[N_\downarrow(E_F)]$ is the density of states (DOS) of up [down] spins at the Fermi level. This quantity is not, however, very useful in transport experiments where the polarization of the injected current depends on the difference in the total flux of spin-up and spin-down electrons which depends both on the DOS and Fermi velocities of the up and down electrons. In a ballistic point-contact experiment, where the electron is allowed to flow from the ferromagnet to another metal/superconductor through an orifice smaller than the mean free path of the carriers (l), the net flux of the carriers with spin σ from a particular band i is given by[6] $\langle N_{i\sigma k} v_{in\sigma k}\rangle$, where $\mathbf{k}$ is the wave vector on the Fermi surface, $\mathbf{n}$ is the direction of the current flow, and the average is taken over the Fermi surface. This quantity can be easily seen to be proportional to $S_{i\sigma\mathbf{n}}$, the area of projection of the ith band with spin σ on the interface plane. Summing over all the bands, the total flux of carriers with spin σ is given by $S_{\sigma\mathbf{n}}$, the total area of projection of the bands with spin σ on the interface plane. Thus the spin polarization of the injected current, commonly known as the transport spin polarization, is given by $P_t=[(S_{\uparrow\mathbf{n}}-S_{\downarrow\mathbf{n}})/(S_{\uparrow\mathbf{n}}+S_{\downarrow\mathbf{n}})]$. $P_t \approx P$, only in an isotropic Fermi surface if the velocities of the up and down spins are equal. Knowledge of P_t is directly relevant in interpreting results in experiments involving the transport of spin-polarized carriers from a ferromagnet across an interface.

The $4d$ itinerant ferromagnet $SrRuO_3$ is an interesting material to explore from this point of view. $SrRuO_3$ stabilizes in an orthorhombically distorted perovskite structure $a \approx b \approx c/\sqrt{2}$. It orders ferromagnetically[7] below 160 K and has a saturation moment of $1.6\mu_B$, which is so far the largest known in any $4d$ ferromagnet.[8] $SrRuO_3$ is thus close to a half metal though the spin polarization at the Fermi level has been predicted to be in the range $P=0.091-0.2$.[9,10] The Fermi velocity of the minority-spin carrier band has, however, been predicted to be 2–3 times larger than the majority band. This should give rise to a large negative transport spin polarization. In the family of oxide ferromagnets, the main attraction of $SrRuO_3$ stems from the fact that it is a ''clean'' system without any substitutional disorder. This, combined with the ability to grow very-high-quality single-crystalline films makes it possible to realize large mean free paths (l ~ 500 Å) in clean samples.[11] The clean ballistic limit which is often difficult to attain in doped systems like hole-doped rare-earth manganites can therefore be reached in a point-contact experiment in this system.

In this paper we report the transport spin polarization in $SrRuO_3$ from Andreev reflection[12] data using a Nb point contact in the ballistic limit. The two films ($\sim$2000 Å) used in this study were grown in a way similar to the films used earlier for quantum oscillations studies.[13] Conductance versus voltage (G-V) characteristics of the Nb-$SrRuO_3$ point contact were measured in the temperature range 2.6–4.2 K

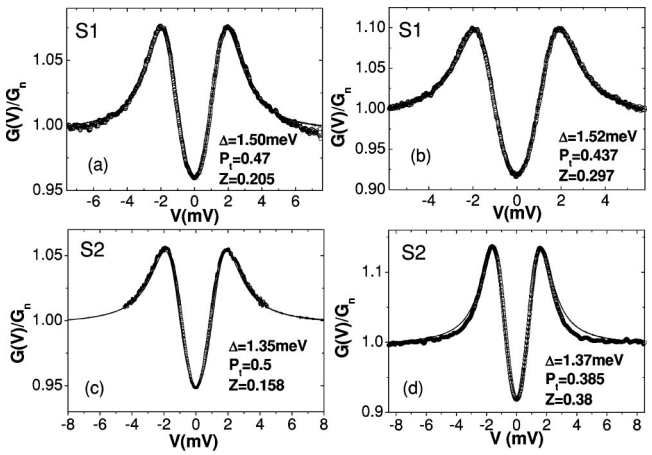

FIG. 1. (a, b) Representative $G(V)/G_N$ vs V plots for the $SrRuO_3$/Nb point contact measured at 4.2 K for $S1$; (c, d) the same for $S2$. The solid lines are the fits to the BTK model. The measured value of P_t decreases from the intrinsic value for dirtier contacts, i.e., larger Z.

by dipping the sample and tip in liquid He and pumping over the He bath. The Nb tip was fabricated either through electrochemical etching of Nb wire in potassium hydroxide solution or by mechanically polishing the tip. No significant difference was observed between the two different kinds of tips. The G-V characteristics of the point contact were measured directly using a modulation technique and averaged over ten sweeps for every G-V curve.

The residual resistivities of the two films used in this study were 6.8 $\mu\Omega$ cm ($S1$) and 5.6 $\mu\Omega$ cm ($S2$). Estimating the electronic mean free path (l) corresponding to these values is not trivial, but evidence from previous studies[11,13] suggests values of several hundred angstroms. The diameter of the point contact (d) can be estimated from the normal-state resistance (R_N) of the point contact using the approximate formula given by Wexler,[14]

$$R_N \approx \frac{4}{3\pi}\frac{\rho l}{d^2} + \frac{\rho}{2d}, \tag{1}$$

where ρ is the resistivity of the sample. For the cleanest point contacts on both $S1$ and $S2$, $R_N \sim 12\ \Omega$. This corresponds to $d \sim 100$ Å. Thus we expect our measurements of transport spin polarization to be well in the ballistic limit ($d < l$) of the point contact. An additional point to note is that the absolute resistance of both the films below 10 K was $\sim 0.5\ \Omega$. Correspondingly, only point contacts with point-contact resistance larger than 10 Ω were analyzed, to avoid any significant voltage contribution to the point-contact spectra from the voltage drop in the sample.

In Figs. 1(a)–1(d) we show four representative plots of the normalized conductance versus voltage of the Nb-$SrRuO_3$ point contact taken at 4.2 K. The different spectra were recorded by engaging the point contact several times on the films. These point contacts differ in the value of the scattering barrier at the interface between the ferromagnet and superconductor, which depends on the microscopic details of the interface. To analyze the G-V characteristic of the point contact we use a modified Blonder-Tinkham-Klapwijk[15] (BTK) scheme, where the total current is decomposed into an unpolarized component and a fully polarized component.[16,17] There has been some controversy in the existing literature regarding the exact form of the reflection and transmission coefficients to be used for the analysis.[18] We are of the opinion that the approach of Ref. 16 is the correct one and use it in our work. For the sake of completeness we briefly outline the model here. We consider the total current across our point contact to be made of an unpolarized component and a fully polarized component. In the model developed by BTK the current (I) is given terms of the Andreev reflection probability $A(E)$ and the normal reflection probability $B(E)$ of an incident electron on the ferromagnet superconductor interface as

$$I \propto N v_F \int_{-\infty}^{\infty} [f(E-eV,T) - f(E,T)][1 + A(E) - B(E)]dE$$
$$= N v_F I'. \tag{2}$$

Here N is the density of states in the ferromagnet and v_F is the Fermi velocity. The coefficients $A(E)$ and $B(E)$ are different for the unpolarized current and the fully polarized current. Since in a typical Andreev spectrum the normalized conductance $G(V)/G_N$ $[\equiv (dI/dV)/(dI/dV)_{eV \gg \Delta}]$ as a function of voltage is fitted, it is enough to evaluate the integral I' without considering the prefactor Nv_F explicitly. For the unpolarized case BTK solved the Bogoliubov–de Gennes[11] (BdG) equations at the interface to find the coefficients $A(E)$ and $B(E)$. An incident particle on the interface (at $x=0$) given by $\Psi_{\rm inc} = \binom{e^{ikx}}{0}$ produces a reflected component $\Psi_{\rm refl} = b\binom{e^{-ikx}}{0} + a\binom{0}{e^{ikx}}$ and a transmitted component $\Psi_{\rm trans} = c\binom{u^s}{v^s}e^{iq_{u^s}x} + d\binom{v^s}{u^s}e^{-iq_{v^s}x}$. Here u^s and v^s are obtained from the solution of the BdG equation in the superconductor: $(u^s)^2 = 1 - (v^s)^2 = (1/2)[1 + \{(E^2 - \Delta^2)/E^2\}^{1/2}]$. The first and second terms in $\Psi_{\rm refl}$ correspond to the normal and Andreev reflection processes, respectively. The coefficients a, b, c, and d are calculated from the boundary conditions

$$\text{(i)} \quad \Psi_n(x=0) = \Psi_s(x=0),$$

$$\text{(ii)} \quad \Psi_s'(x=0) - \Psi_n'(x=0) = \frac{2mV_0}{\hbar^2}\Psi(x=0),$$

where $\Psi_n(x) = \Psi_{\rm inc}(x) + \Psi_{\rm refl}(x)$ and $\Psi_s(x) = \Psi_{\rm trans}(x)$ are the wave functions inside the normal metal and superconductor, respectively. The interfacial scattering at the ferromagnet/superconductor interface (originating both from interfacial scattering from an imperfect junction such as an oxide barrier and from the mismatch in the Fermi energies between the metal and superconductor[19]) is simulated through a δ-function potential of the form $V_s(x) = V_0\delta(x)$ at the interface. The coefficients $A(E)$ and $B(E)$ correspond to the probability currents associated with the Andreev and normal reflection processes and are given by $A(E) = a^*a$ and $B(E) = b^*b$. In the fully polarized scenario, the allowed k vectors are only in one spin direction and the Andreev-reflected hole cannot propagate since it has a spin opposite to the incident electron. Therefore the Andreev-reflected com-

TABLE I. The coefficients A(E) and B(E) corresponding to the polarized and unpolarized cases. $\gamma^2=\{[(u^s)^2-(v^s)^2]Z^2+(u^s)^2\}^2$, $Z=V_0/\hbar v_F$ and $\epsilon=[(E^2-\Delta^2)/E^2]$. The subscripts u and p denote the coefficients for the unpolarized and polarized current, respectively.

	$E<\Delta$	$E>\Delta$
$A_u(E)$	$\frac{(\Delta/E)^2}{1-\epsilon(1+2Z^2)^2}$	$\frac{(u^s v^s)^2}{\gamma^2}$
$B_u(E)$	1-$A(E)$	$\frac{[(u^s)^2]-[(v^s)^2]^2Z^2(1+Z^2)}{\gamma^2}$
$A_p(E)$	0	0
$B_p(E)$	1	$\frac{(\epsilon^{1/2}-1)^2+4Z^2\epsilon}{(\epsilon^{1/2}+1)^2+4Z^2\epsilon}$

ponent gives rise to an evanescent wave. In this case the reflected component is given by $\Psi_{\rm refl}=b\binom{e^{-ikx}}{0}+a\binom{0}{e^{\kappa x}}$, where κ is inversely proportional to the decay length of the evanescent wave. The evanescent wave does not carry any current so $A(E)=0$. $B(E)=b^*b$ can be calculated using the same boundary conditions as before. In Table I we list the $A(E)$ and $B(E)$ for the polarized and unpolarized cases assuming $\kappa\rightarrow\infty$.[16] For an arbitrary transport polarization P_t the total current will be given by

$$I=I_u(1-P_t)+I_pP_t, \tag{3}$$

where I_u and I_p are given by Eq. (1) using the unpolarized and polarized $A(E)$ and $B(E)$ coefficients, respectively. The experimental point-contact spectra are fitted [solid line in Figs. 1(a)–1(d)] with the strength of the scattering barrier, Z $(=V_0/\hbar v_F)$, the transport spin polarization P_t, and the superconducting energy gap Δ as fitting parameters.[20] We believe that the difference between the fitted values of Δ for the point contacts on $S1$ and $S2$ arises from variation in the quality of tips produced by our tip fabrication process. Note, however, the consistency of the value fitted from different spectra using the same tip and that (as expected) none of the values exceed that of bulk Nb. The value of P_t extracted from the fits decreases with increasing Z, the scattering strength at the interface. This behavior has been observed earlier in iron, cobalt, and nickel films as well as CrO_2 and in manganites.[17,21] It is believed to be due to the spin mixing effect at the magnetically disordered scattering barrier formed at the interfaces.[22,23] Andreev reflection therefore provides a lower bound on the transport spin polarization.

In order to extract the intrinsic spin polarization we plot (Fig. 2) P_t as a function of Z obtained from the Andreev spectra at 4.21 K from both samples $S1$ and $S2$. Within experimental errors, we could fit our Z dependence of P_t with a parabolic curve for data points obtained from both $S1$ and $S2$. Thus we extract P_t in the limit $Z\rightarrow 0$ by extrapolating back a fitted parabolic curve. The intrinsic polarization obtained from the fit is $P_t=0.51\pm 0.02$.

The fact that data points obtained from both $S1$ and $S2$ can be fitted with a single parabolic curve giving the same value for P_t though their residual resistances are different is important. Mazin *et al.*[16] have pointed out that in the diffusive limit of a point contact $(d\gg l)$ the transport spin polarization is given by $P_t=(\langle N_\uparrow v_{F\uparrow}^2\rangle-\langle N_\downarrow v_{F\downarrow}^2\rangle)/(\langle N_\uparrow v_{F\uparrow}^2\rangle+\langle N_\downarrow v_{F\uparrow}^2\rangle)$ instead of $P_t=(\langle N_\uparrow v_{F\uparrow}\rangle-\langle N_\downarrow v_{F\downarrow}\rangle)/(\langle N_\uparrow v_{F\uparrow}\rangle+\langle N_\downarrow v_{F\uparrow}\rangle)$. Since these two quantities are normally different for a ferromagnet in a borderline case, when $d\approx l$, one can see a systematic change in the measured value of P_t, as a function of sample disorder.[24] The unique value of the spin polarization obtained for both films confirms that our measurements are well in the ballistic limit $(d\ll l)$ of the point contact. As a further check on our experiment and data analysis procedure, we studied the temperature dependence for the point contact with lowest Z on $S1$. As expected, the extracted values of P_t $(=0.48\pm 0.02)$ extracted from the spectra are constant within error bars over this temperature range.

It is interesting to note that the value of P_t measured in this study is much larger than the spin polarization P predicted from band structure calculations.[9,10] Though the two band structure calculations so far published on this compound differ in their detail, the predicted value of P is small: $P=0.091$ (Ref. 10) and $P=0.2$ (Ref. 9), respectively. The large value of P_t compared to the value of the spin polarization P predicted from band structure calculations suggests that the large transport spin polarization originates primarily from a difference in the Fermi velocities of minority- and majority-spin bands.[25] This is also in agreement with both band structure calculations which predict the average Fermi velocity of the carriers in the majority-spin band to be smaller by a factor of 2–3 than those in the minority-spin band.

In this context it is also interesting to compare our results with the spin polarization measured by Worledge and Geballe using the Meservey-Tedrow technique.[26] Within a simple model the spin polarization measured with this technique is similar to that given by Andreev reflection in the diffusive limit. The main advantage of this technique over

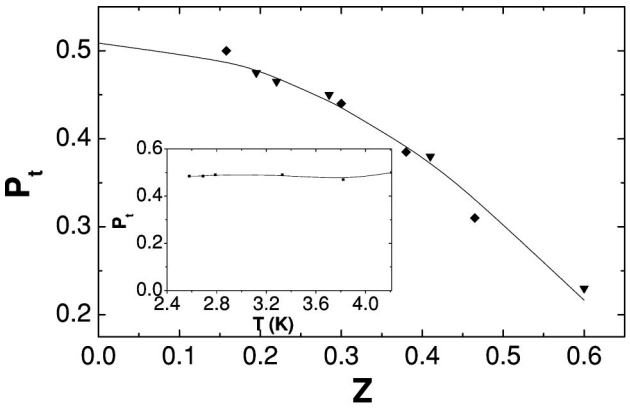

FIG. 2. P_t as a function of Z for different point contacts. The triangles and diamonds on the P_t vs Z plot are data points taken from samples $S1$ and $S2$, respectively. The dashed line is a parabolic fit to the data to extract the transport spin polarization for $Z=0$. The inset shows the transport spin polarization for $S1$ measured at different temperatures. The solid line is a guide to the eye.

Andreev reflection is that it is also sensitive to the sign of the spin polarization. Consistent with band structure predictions, Worledge and Geballe observed a negative transport spin polarization in $SrRuO_3$. However, the degree of spin polarization obtained by them (~9.5%) is much smaller than the transport spin polarization expected from band structure calculations. The main disadvantage of the Meservey-Tedrow technique is that the measured transport spin polarization depends on the spin decay length in the tunnel barrier.[26] Therefore the degree of spin polarization measured with this technique depends on the insulating spacer material used and may not reflect the true spin polarization in the material. Andreev reflection in the clean limit, on the other hand, does not suffer from this drawback.

Finally, we would like to note that most spin injection experiments in *d*-wave superconductors have used doped rare-earth manganites such as $La_{0.7}Sr_{0.3}MnO_3$ or $La_{0.7}Ca_{0.3}MnO_3$ as the natural choice, in view of their half-metallic character observed from spin-polarized photoemission experiments.[27] However, in recent times several experiments have cast doubt on the use of this material as the ideal choice. Point-contact Andreev reflection measurements by Nadgomy *et al.*[24] on $La_{0.7}Sr_{0.3}MnO_3$ showed the polarization to be much less than 100%. Measurements of spin polarization by Ji *et al.*[28] using the same technique in the diffusive limit on single crystals of $La_{0.7}Sr_{0.3}MnO_3$ and $La_{0.6}Sr_{0.4}MnO_3$ support these results. Spin-polarized photoemission also showed that the spin polarization of the surface layer decays much more rapidly with temperature than the bulk.[29] It is therefore interesting to note that the transport spin polarization of $SrRuO_3$ in the ballistic limit is comparable to $La_{0.7}Sr_{0.3}MnO_3$. The advantage of $SrRuO_3$ is that high-quality thin films of this material have much lower residual resistance owing to the absence of substitutional disorder and the ability to grow very-high-quality single-crystalline films, thereby reducing the problems associated with Joule heating in spin injection experiments. It would therefore be interesting to compare the effect of spin injection from $SrRuO_3$ with the data from doped manganites.

In summary, we have measured the transport spin polarization in epitaxial thin films of $SrRuO_3$ in the ballistic limit. The transport spin polarization in this compound is comparable to the doped rare-earth manganites. Comparing with the value of the spin polarization calculated from band structure predictions, we conclude that the large transport spin polarization is mainly due to the difference in the Fermi velocities of the majority- and minority-spin carrier electrons. This agrees well with the prediction from band structure calculations.

We would like to thank the Leverhulme Trust and the Royal Society for research grants and G. R. Walsh and D. A. Brewster for help with constructing the experimental apparatus and S. A. Grigera for critically reading the manuscript. We are also grateful to M. S. Osofsky for sharing his group's results prior to publication.

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