Measurement of spin polarization of single crystals of $La_{0.7}Sr_{0.3}MnO_3$ and $La_{0.6}Sr_{0.4}MnO_3$

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The spin polarization of single crystals $La_{0.7}Sr_{0.3}MnO_3$ and $La_{0.6}Sr_{0.4}MnO_3$ has been measured by point-contact Andreev reflection (PCAR) using a superconducting Pb tip. The conductance vs bias voltage spectra have been taken at temperatures below 2 K for different contact junction resistances. The quadratic dependence of polarization upon scattering barrier strength, similar to those of other ferromagnetic materials measured by PCAR, has been established. The intrinsic values of polarization have been determined to be 0.78 ± 0.02 and 0.83 ± 0.02 for $La_{0.7}Sr_{0.3}MnO_3$ and $La_{0.6}Sr_{0.4}MnO_3$, respectively. The possibility of interfacial and bulk diffuse scattering has been discussed and both tend to dilute the intrinsic spin polarization.

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The strontium-substituted lanthanum manganites, such as $La_{1-x}Sr_xMnO_3$ (LSMO), have attracted a lot of attention in recent years due to their intriguing physics and colossal magnetoresistance. Another interesting property of this material is the value of spin polarization, which is usually defined as the imbalance of the density of states for two spin orientations at the Fermi energy. The performance of future magnetoelectronic devices depends critically on materials with high spin polarization.[1,2] Theoretically, $La_{0.7}Sr_{0.3}MnO_3$ has been predicted to be highly spin polarized, and perhaps half-metallic with 100% spin polarization.[3] Spin-resolved photoemission results of LSMO by Park *et al*.[4] have reported on the half-metallic nature, however, superconducting tunnel junction measurement of LSMO/$SrTiO_3$/Al has indicated a polarization of only 0.72.[5]

Recently, it has been shown that point-contact Andreev reflection (PCAR) with suitable analyses of the conductance results can accurately measure the spin polarization of a ferromagnet.[6–8] Using PCAR, CrO_2 has been demonstrated to be half-metallic with a polarization no less than 0.96.[9] It was also realized that a surface oxidation/degradation layer could significantly dilute the bulk value of the spin polarization of the material.[9,10] LSMO has been studied by this technique in previous works.[7,11,12] But a systematic study, taking into account the effects of the interfacial scattering and bulk scattering, is still lacking. In this work, we present the PCAR measurement on single crystals of LSMO.

The single crystals of $La_{0.7}Sr_{0.3}MnO_3$ were grown by the floating-zone method.[13] The crystal is usually regarded to have a pseudocubic lattice structure. The x-ray θ-2θ scans for (001), (011), and (111) planes of a $La_{0.7}Sr_{0.3}MnO_3$ crystal are shown in Figs. 1(a)–1(c), respectively. The lattice constant of the pseudocubic structure has been determined to be 3.88 Å. Figures 1(d)–1(f) show the rocking curves of the strongest peak for each crystal plane. The rocking curves for (011) and (222) peaks have full widths at half maximum (FWHM) of 0.02° and 0.04°, respectively, whereas the rocking curve for (002) has three prominent peaks. Nevertheless, each individual peak is very narrow with a FWHM no more than 0.03° and the overall span of the three peaks is less than 0.3°. These x-ray data demonstrate the high quality of the single-crystal LSMO.

The temperature dependence of magnetization of the $La_{0.7}Sr_{0.3}MnO_3$ crystal [Fig. 2(a)] was measured in an external field of 1.5 T, showing a Curie temperature of about 380 K. The inset of Fig. 2(a) shows a hysteresis loop taken at 5 K. The magnetization is plotted in units of Bohr magnetons (μ_B) per lattice site (per Mn site). The magnetization at 5 K is found to be $3.5\mu_B$. Figure 2(b) shows the resistivity as a function of temperature. The resistivity decreases by two orders of magnitude from T_C to 5 K. The inset of Fig. 2(b) shows the resistivity data below 40 K. The residual resistivity of the material is about 45 $\mu\Omega$ cm.

The PCAR measurements were carried out in a manner described elsewhere.[9,10] Andreev reflection is the process of quasiparticle-current-to-supercurrent conversion at the metal/superconductor interface. The PCAR technique utilized the fact that the imbalance of the density of states for two spin orientations at the Fermi energy of a ferromagnet reduces the probability of Andreev reflection. In the case of transparent contact without interfacial scattering, the normalized conductance spectra are related to the spin polarization in a simple manner $G(0)/G_n=2(1-P)$, where $G(0)$ is the conductance at zero-bias voltage, G_n is the conductance in the normal state, and P is the polarization of the ferromagnet.[7] In general, a modified version[10] of Blonder-Tinkham-Klapwijk (BTK) theory[14] should be used to analyze the spectra in order to obtain the value of spin polarization. Note that, in contrast to the usual definition of spin polarization by the density of states, PCAR measures the polarization of the spin currents.[7–10]

In this work, all the PCAR measurements were done below 2 K with a superconducting Pb tip. The tip was repeatedly brought into contact with the surface of the bulk single crystal, and the conductance vs bias voltage spectra were taken for various contact resistances. Figure 3 shows six rep-

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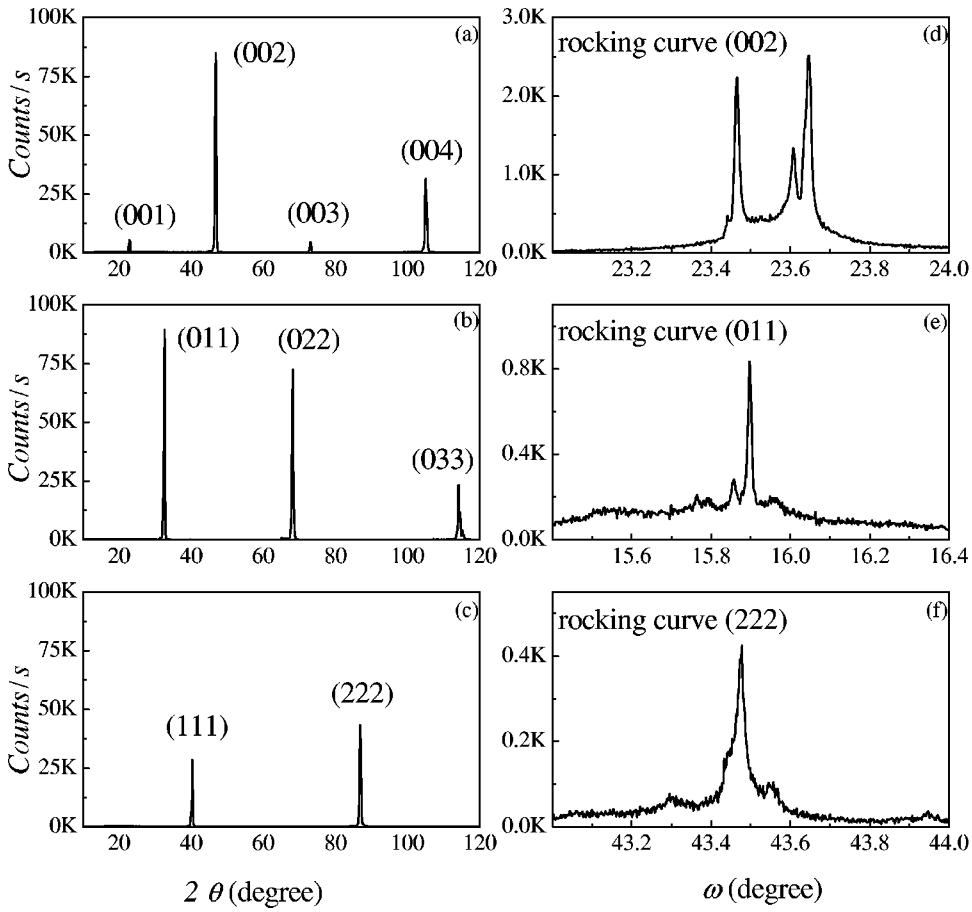

FIG. 1. θ-2θ diffraction of (a) (001), (b) (011), and (c) (111) planes of a single-crystal $La_{0.7}Sr_{0.3}MnO_3$. Rocking curves for (d) (002), (e) (011), and (f) (222) peaks.

resentative conductance curves (open circles) and fits using the modified BTK theory (solid lines). Spin polarization P, interfacial scattering barrier Z, and superconducting energy gap Δ were used as the fitting parameters. All the fitting parameters, temperatures for the measurements, and the contact resistances are shown in the figures for each spectrum. In all the spectra, there is a deep trench at low bias voltage and two peaks, with different heights, at a voltage near the gap value of the superconductor. The deep trench at low bias voltage with significantly reduced zero-bias conductance in the spectra is mostly due to the high degree of spin polarization in the LSMO and to a lesser extent the interfacial scattering which is characterized by the Z factor. The higher the Z value, the higher the peaks at the gap value in the spectra. The spectra in Figs. 3(a)–3(f) are arranged in a decreasing order of the fitted Z factors. The Z values range from about 1.0 to 0.17. In Figs. 3(e)–3(f), the Z values are 0.27 and 0.17, respectively, which represent very transparent interfaces. The reduced values of $G(0)/G_n$ are due to spin polarization, not interfacial scattering. When a clean interface with $Z=0$ is achieved, the spin polarization in the spectrum can be obtained by the simple relation of $G(0)/G_n = 2(1-P)$. It is instructive to compare the spectra of Figs. 3(a) and 3(f). In Fig. 3(a), the zero-bias conductance value is lower than that of Fig. 3(f). But the spin-polarization value of the spectrum in Fig. 3(a) (0.53) is in fact much lower than that in Fig. 3(f) (0.77). This observation underscores the importance of quantitative analyses of the PCAR results.

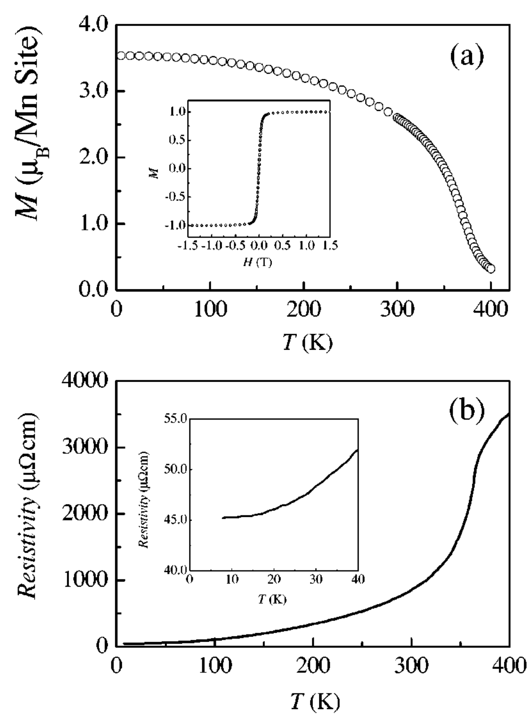

FIG. 2. Temperature dependence of (a) magnetization at a field of 1.5 T and (b) resistivity of a $La_{0.7}Sr_{0.3}MnO_3$ crystal. The hysteresis loop at 5 K is shown in the inset of (a), and the resistivity at low temperatures is shown in the inset of (b).

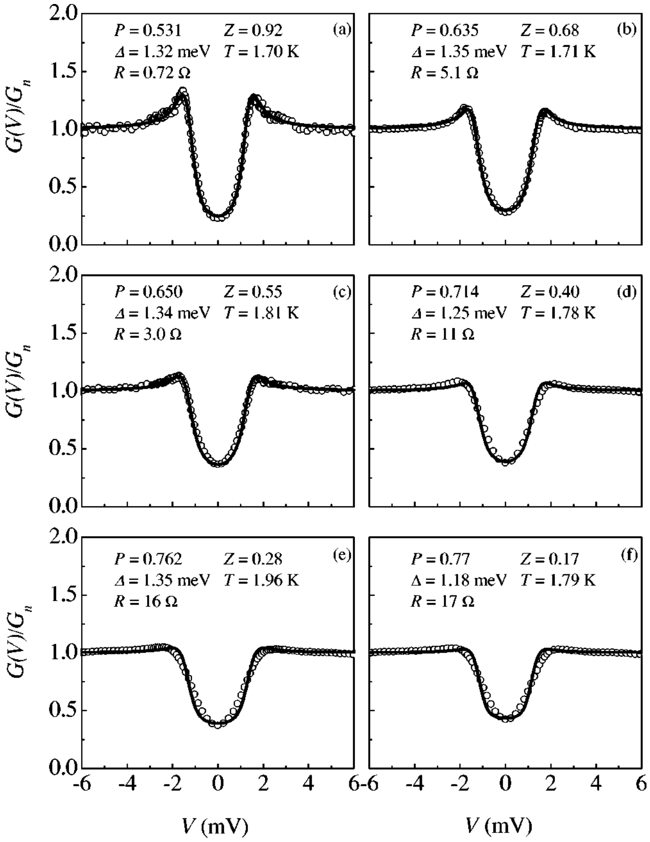

FIG. 3. Various normalized conductance vs bias voltage curves (open circles) of a $La_{0.7}Sr_{0.3}MnO_3$ crystal and fits (solid lines) using the modified BTK model.

In Fig. 4, the fitted spin-polarization values of each spectrum as a function of Z are plotted as open squares. The dependence of P upon Z is obvious and consistent with our previous works[9,10] on other materials: spectra with higher barrier strength tend to have lower spin polarization. The solid line in the figure is a quadratic fit to the data. The intrinsic value of spin polarization of single-crystal $La_{0.7}Sr_{0.3}MnO_3$ has been determined as 0.78 ± 0.02 by extrapolating the solid curve to $Z=0$.

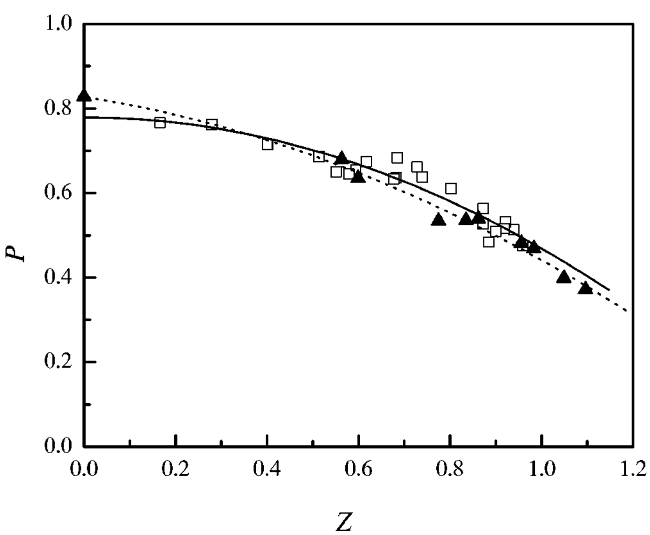

FIG. 4. Measured spin polarization P as a function of barrier strength Z of $La_{0.7}Sr_{0.3}MnO_3$ (open squares) and $La_{0.6}Sr_{0.4}MnO_3$ (solid triangles) crystals. The solid and dashed lines are quadratic fits to the data of $La_{0.7}Sr_{0.3}MnO_3$ and $La_{0.6}Sr_{0.4}MnO_3$, respectively.

Some of the spectra with high barrier strength have low contact resistance, for example, in Fig. 3(a). We have measured junctions with resistance as low as 0.1 Ω and still observed a Z close to 1. Considering the relatively high resistivity of this material, these low ohmic junctions could be in the diffusive regime. The residue resistivity $\rho=45\ \mu\Omega$ cm and the carrier (hole) density n are found to be $1.5\times10^{22}\ \mathrm{cm}^3$ by the Hall effect.[15] Using these two parameters, the carrier mean free path can be estimated from the Drude model to be $l=46$ Å. The expression for the normal-state contact resistance, $R_n=4\rho l/3\pi a^2+\rho/2a$, can be used to estimate the contact size a.[16] For $R_n=17\ \Omega$, as in Fig. 3(f), a is about 160 Å. For $R_n=0.72\ \Omega$, as in Fig. 3(a), a is about 3000 Å. Thus, in all the junctions, the junction size is larger than the carrier mean free path. The observed finite Z behavior of all the junctions could be a combination of interfacial scattering and scattering associated with diffusive transport. The two scattering mechanisms may not be distinguishable from each other by the measured spectra but both can be distinguished from the effect of high spin polarization.[17] The existence of diffusive bulk scattering merely increases the effective Z factor of the spectra. The high Z factor observed in Fig. 3(a) is very likely due to diffusive bulk scattering, since the estimated contact size a (3000 Å) is much larger than the mean free path (46 Å). Though the estimated contact size a for the results in Figs. 3(e) and 3(f) (160 Å) is larger than mean free path l (46 Å), they are roughly on the same order of magnitude. The low Z behavior of the spectra in Figs. 3(e) and 3(f) demonstrates a transparent interface and minimal bulk scattering, and the contact junctions are arguably near the ballistic limit. Most importantly, as we can appreciate from the quadratic dependence of P upon Z in Fig. 4, bulk scattering in the diffusive regime also tends to dilute the intrinsic spin polarization of the material just as the interfacial scattering does. This is because spin-flip processes exist in both scattering mechanisms. It is also noted that in Figs. 3(e) and 3(f) there is a slight but noticeable discrepancy between the data and the fitted curves. The seemingly extra broadening of the data can be explained by a finite quasiparticle lifetime[18] and the proximity effect.

The same type of PCAR measurements have also been applied to another single crystal of composition $La_{0.6}Sr_{0.4}MnO_3$. We have obtained a similar P vs Z dependence shown as solid triangles in Fig. 4. The dotted line is a quadratic fit to the data and an intrinsic spin polarization of 0.83 is determined in the limit of $Z=0$.

The intrinsic spin polarization of 0.83 for $La_{0.6}Sr_{0.4}MnO_3$ is similar to the value of 0.78 measured for $La_{0.7}Sr_{0.3}MnO_3$. In both cases, the spin polarization of LSMO is very high, second only to CrO_2, which has a spin polarization of 96%–98.4%.[9,19] However, we have found no evidence that LSMO is half-metallic, despite theoretical predictions[3] and experimental claims.[4] The main experimental claim of half-metallicity of LSMO is the spin-resolved photoemission results of Park *et al*.[4] It is well known that photoemission is highly surface sensitive with contributions mainly from the top 5–10 Å of the material, which may be different from the bulk material. On the other hand, polarization measured by Andreev reflection and superconducting tunnel junction relies on spin-polarized current across the interface.

We note, however, that one of the signatures of half-metallicity is the integer magnetic moment per unit cell. The total number of electrons per unit cell $n = n_{\uparrow} + n_{\downarrow}$ is an integer, where $n_{\uparrow}$ and $n_{\downarrow}$ are the number of electrons per unit cell for spin up and spin down, respectively. For a half-metal, one spin band is completely filled. This leads to the situation where $n_{\uparrow}$, $n_{\downarrow}$, and $n_{\uparrow} - n_{\downarrow}$ must all be integers. The net magnetization is $\mu_B(n_{\uparrow} - n_{\downarrow})$. Thus, one of the necessary but not sufficient conditions for a half-metal is that it has an integer magnetic moment per unit cell. For example, one observes $2\mu_B$ in CrO_2. In the case of $La_{0.7}Sr_{0.3}MnO_3$, an extension of the criterion of integer magnetic moment is necessary to accommodate solid solutions.[20] We have measured a moment of $3.5\mu_B$, which is less than the $3.7\mu_B$ expected for a half-metal of this particular composition.[21] From the literature, the magnetic moment per Mn site of the so-called optimally doped LSMO is consistently less than $3.7\mu_B$.[21] From this point, we argue that LSMO is unlikely to be a half-metal, contrary to some theoretical predictions[3] and experimental observation.[4] It is nevertheless a metal with a very high spin polarization.

In conclusion, we have measured the spin polarization of $La_{0.7}Sr_{0.3}MnO_3$ and $La_{0.6}Sr_{0.4}MnO_3$ by the PCAR technique and we have found the intrinsic values of polarization to be 0.78 ± 0.02 and 0.83 ± 0.02, respectively. Despite very high spin polarization, these materials are not half-metals. In the PCAR measurements, both interfacial scattering and bulk diffusive scattering (if any) tend to dilute the intrinsic spin polarization.

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