Reliability of normal-state current–voltage characteristics as an indicator of tunnel-junction barrier quality

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(Received 21 April 2000; accepted for publication 26 July 2000)

We demonstrate that one of the most commonly used criteria to ascertain that tunneling is the dominant conduction mechanism in magnetic tunnel junctions—fits of current–voltage (I–V) data—is far from reliable. Using a superconducting electrode and measuring the differential conductance below T_c, we divide samples into junctions with an integral barrier and junctions having metallic shorts through the barrier. Despite the clear difference in barrier quality, equally reasonable fits to the I–V data are obtained above T_c. Our results further suggest that the temperature dependence of the zero-bias resistance is a more solid criterion, which could therefore be used to rule out possible pinholes in the barrier.
[S0003-6951(00)01138-4]

Ferromagnet/insulator/ferromagnet ($F/I/F$) tunneling structures exhibit large magnetoresistance (MR)[1] which makes them attractive for magnetic-field sensors[2] and nonvolatile memory devices.[3] Since the tunneling current can exhibit large spatial inhomogeneity even for seemingly perfectly flat barriers,[4] it is possible that for small barrier thickness pinholes will appear and effectively short the tunneling current. Recently, García and co-workers studied Ni–Ni (Ref. 5) and Co–Co (Ref. 6) wire nanocontacts, observing MR values close to 300% at room temperature and 100 Oe applied field. This raises the intriguing question of whether pinholes contribute to the large MR values observed in magnetic tunnel junctions (MTJs). The existence of a high density of pinholes contributing MR would represent a problem for reproducibility when downscaling MTJs to lateral dimensions comparable to the average pinhole separation.

As electron tunneling is known to be ''an unlikely mechanism of current flow through an insulator,''[7] and other conduction paths can contribute significantly, the ''Rowell criteria''[8] were formulated to ascertain that single-step tunneling is the dominant conduction mechanism in junctions with at least one superconducting electrode. For $F/I/F$ structures three tests remain: (i) an exponential insulator thickness (t) dependence of the conductance,[9] $G(t)\sim\exp(-t/t_0)$, with $t_0=\hbar/2\sqrt{2m\phi}$, (ii) a parabolic voltage (V) dependence of the conductance $G(V)$ that can be fitted to theoretical models,[10,11] and (iii) a weak insulating-like temperature dependence $G(T)$.

A reasonable barrier-growth model, where insulating material is randomly deposited onto a metallic surface, leads (as the large-sample limit of the Poisson distribution) to an exponential probability distribution for pinholes through the barrier.[12] Although based entirely on a classical conduction mechanism, the conductance of such a system will therefore also exhibit an exponential thickness dependence, $\exp(-t/t_0)$, where t_0 is approximately 1 monolayer, very close to the Wentzel–Kramers–Brillouin decay length above. (i) is hence a necessary, but not a sufficient, condition for establishing that tunneling is the dominant conduction mechanism.

The second criterion is more prevalent in the literature on $F/I/F$ trilayers as a ''proof'' of tunneling. Fitting the experimental current–voltage relation (I–V) to a theoretical expression, most often of the Simmons type,[10] provides the barrier height ϕ and its thickness d, or in the case of the Brinkman–Dynes–Rowell model,[11] also yields the barrier asymmetry $\Delta\phi$. If ''reasonable'' values are obtained, one concludes that tunneling dominates the conduction.

In this letter we show that these fits do not rule out the existence of pinholes in the insulating barrier. In superconductor/insulator/ferromagnetic ($S/I/F$) structures, we uniquely establish the two types of conduction mechanisms below the superconducting transition temperature T_c: tunneling by the observation of a Bardeen–Cooper–Schrieffer type density of states, and pinhole conduction by the observation of Andreev reflection. Irrespective of the dominant conduction mechanism, fits to both models at $T>T_c$ provide reasonable barrier parameters.

We also investigate the third criterion and find a clear correlation between insulating-like $G(T)$ and tunneling on the one hand, and metal-like $G(T)$ and pinholes on the other. Out of the three Rowell criteria applicable to $F/I/F$ trilayers only one, the temperature dependence of the conductance, seems reliable.

Using dc sputtering, thermal, and e-beam evaporation deposition, combined with barrier oxidation in air and/or oxygen glow discharge, we prepared over 500 trial junctions with different $S/I/F$ combinations. From G–V measurements at room temperature we divided samples into two categories: (1) samples exhibiting nonlinear G–V characteristics (possible junctions), and (2) samples with no bias

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dependence, which were not considered for further studies (obvious contacts). The number of promising samples was further reduced by requiring their resistance to fall between 10 and 1000 Ω. Out of the 16 samples that were studied in great detail we present data on two characteristic samples, showing good tunneling and contact properties, respectively.

Both samples have the same structure: a dc sputtered superconducting Nb/Al bilayer bottom electrode, an insulating aluminum oxide barrier, and a dc sputtered Fe top electrode capped with Al. Nb(70 nm) covered by Al(10 nm) was deposited through a 1 mm shadow mask. SiO_2(100 nm) was rf sputtered on the sides of the bottom electrode to avoid edge effects. After storage in air for several days, the Al layer was glow discharged ($p_{O_2}=350$ mTorr, 350 V dc bias) for 1.5 h to ensure formation of a thick pinhole-free AlO_x barrier. The resulting oxide thickness is typically 2–3 nm and the remaining 8 nm of Al is superconducting from proximity to the Nb.[13] Finally Fe(30 nm) top electrodes were deposited through a 0.3 mm mask followed by 30 nm of Al to prevent oxidation. Sample A has a junction area of 1000 $\times 300\,\mu\mathrm{m}^2$ and a room-temperature resistance–area product RA$=8.2\,\mathrm{M}\Omega\,\mu\mathrm{m}^2$.

For sample B the Nb(80 nm) bottom electrode was covered by Al(10 nm), subsequently exposed to air for 40 min to obtain a thin AlO_x barrier (1–2 nm), and Fe(50 nm) was deposited on top. The Fe layer was photolithographically patterned into squares, the bottom electrode was insulated with SiO_2, and electrical contact was made to the Fe using electrodes patterned from a Al(50 nm) layer. Sample B has a junction area of $50\times 50\,\mu\mathrm{m}^2$ and RA$=0.45\,\mathrm{M}\Omega\,\mu\mathrm{m}^2$ at room temperature.

Standard ac (1 kHz) differential conductance measurements as a function of dc bias were carried out from liquid helium temperatures to room temperature using a conventional balanced bridge. dc bias was limited to $|V|<0.15$ V since some junctions exhibited small but irreversible characteristics changes at higher bias. $I-V$ curves were obtained by numerically integrating the $G-V$ data.

In Fig. 1(a) we show the normal-state $I-V$ curve for sample A together with a fit to Simmons's formula[10] with barrier parameters $d=2.89$ nm and $\phi=0.49$ eV. The inset shows the conductance data for the same sample with a fit to the Brinkman–Dynes–Rowell model yielding essentially the same barrier parameters since the $G-V$ curve is highly symmetric. Fitting the integrated data to Simmons's formula neglects much of the higher sensitivity of the differential conduction measurement, in particular about $V=0$. We choose this representation since it is the common way to test for tunneling in $F/I/F$ structures. Figure 1(b) shows normal-state $I-V$ and conductance data for sample B with fits to Simmons's and the BDR formulae, respectively. Equally reasonable barrier parameters of $d=1.72$ nm and $\phi=0.71$ eV were obtained. The thinner barrier width seems consistent with the shorter oxidation time for sample B. Had these samples been of the $F/I/F$ type we would have concluded that both samples are good tunnel junctions with pinhole-free barriers. However, this conclusion is immediately invalidated when the samples are cooled below T_c of the Nb electrode. Sample A shows a typical signature of tunneling into a superconducting gap at finite T: a *reduced* conductance at $V=0$ and two symmetric maxima just outside $\pm\Delta$ [Fig. 2(a)]. The dI/dV data for sample B are very different and in particular show a 13.7% conductance *increase* in the superconductor gap region [Fig. 2(b)]. This behavior is consistent with Andreev reflection[14] at a highly transmissive (S/N) interface.[15] The ratio of the conductance inside and outside

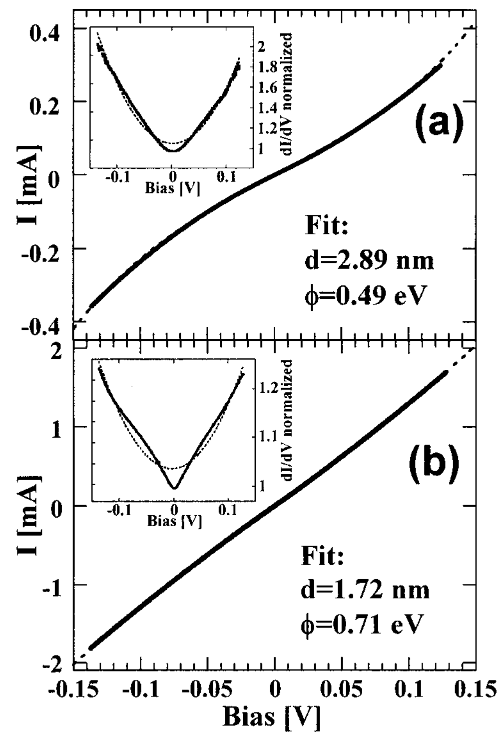

FIG. 1. (a) $I-V$ curve for sample A at $T=90$ K together with a fit to Simmon's model (dashed line). Inset: original conductance data fitted with the BDR model (dashed line). (b) Same for sample B at $T=77$ K.

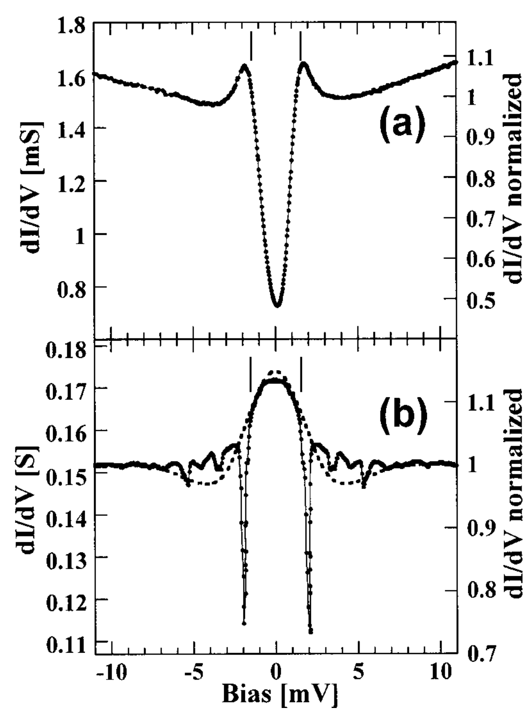

FIG. 2. $G-V$ data for sample A (a) and B (b) taken at $T=4.2$ K. The lines indicate the gap voltage for bulk Nb, ± 1.5 mV. The dashed line in (b) is data from Ref. 17 for an Fe–Ta point contact with the bias scale multiplied by the gap ratio of Nb to Ta.

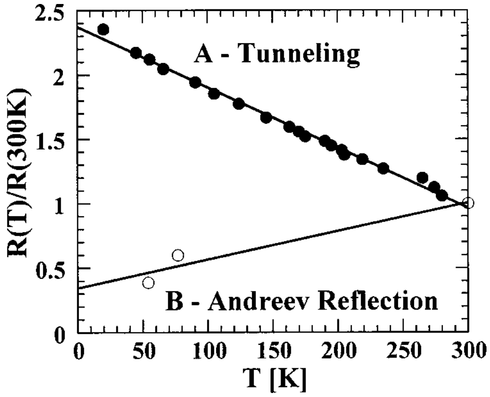

FIG. 3. Zero-bias resistance vs T for samples A (●) and B (○).

of the gap has recently been used as a means to determine the spin polarization of the normal electrode.[16,17] From the data in Fig. 2(b) we get

$$P_{\mathrm{Fe}} = 1 - \frac{1}{2}\frac{G_{V=0}}{G_{\mathrm{normal}}} = 1 - \frac{1.137}{2} = 43\%$$

in excellent agreement with literature values obtained using AR and Zeeman split tunneling spectroscopy data.[17,18] The general shape of the data is similar to that for a Fe–Ta contact, and the dashed line in Fig. 2(b) is data from Ref. 17 with the bias scale multiplied by the gap ratio of Nb to Ta. We also note the presence of sharp resistivity spikes outside the superconducting gap, as is commonly observed for S/N nanocontacts.[19,20] Taken together, these observations conclusively indicate that sample B suffers from a short between the Fe and Nb electrodes. The other samples studied in detail could also be classified as clear tunnel junctions or clear S/F contacts. Equally good fits of the $I-V$ data could be made to Simmons's formula at $T > T_c$.[21] It is hence safe to conclude that a fit above T_c cannot be used as a criterion to ascertain whether or not a tunneling barrier is free of pinholes.

Finally, we turn to the third criterion. The two samples exhibit completely different temperature dependencies of the resistance (Fig. 3). Sample A, being a tunnel junction, shows a weakly insulating behavior, whereas sample B, which contains a short, appears metallic. All other samples that either showed clear tunneling or clear AR correlated completely with these temperature dependencies. We hence conclude that a metallic-like temperature dependence of the zero-bias resistance is incompatible with a pinhole-free barrier. To ensure the integrity of the barrier in $F/I/F$ junctions one therefore should always check that the temperature dependence is insulator-like, even if the $I-V$ data can be fitted with reasonable barrier parameters. The observed combination of a parabolic $G-V$ relation and a metallic-like $G(T)$ dependence most likely arises from a competition between a tunneling path and a contact path. As the temperature is decreased the metallic contact will dominate the conduction and give the observed $G(T)$ as well as the AR below T_c.

In conclusion we have shown that very reasonable fits of the $I-V$ data can be made for tunneling samples that suffer from shorts through the insulator barrier. We have hence clearly demonstrated how unreliable the common fitting criterion is for the determination of the barrier quality. Our data suggest that the temperature dependence of the zero-bias resistance is a much more reliable indicator of an intact barrier. We therefore suggest that the temperature dependence of the junction resistance should always be measured as a check to rule out the presence of pinholes.

The authors thank H. Suhl, P. Prieto, G. Bolaños, and A. Romero for fruitful discussions. R.E. thanks the John Simon Guggenheim Memorial Foundation, and CONACYT-MEXICO. This work was supported by DARPA and ONR.

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