Effect of spontaneous polarization of the ferroelectric substrate on the electrical properties of films

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The effect of a spontaneous polarization of $BaTiO_3$ and $LiNbO_3$ ferroelectric substrates on the electrical properties of $YBa_2Cu_3O_x$ films has been studied. The change in the resistance in the mixed resistive state of the films on the $BaTiO_3$ substrate upon a change in the polarization direction is about 5%. There is a correlation between the change in the resistance and the ferroelectric hysteresis loop. Large changes are found in the electrical properties of films deposited on $LiNbO_3$ substrates with oppositely directed spontaneous polarizations.

The effect of an electric field on the properties of high-T_c superconducting films in superconductor-insulator-metal structures has recently attracted considerable research interest (see, for example, Ref. 1–5 and the papers cited there). It has been suggested that an electric field set up by bound charge in the insulator is screened by free carriers in the superconductor. As a result, there are a curvature of bands and a change in the carrier density in a surface layer with a thickness on the order of the Thomas-Fermi screening length. In a high-T_c superconductor, this dimension is about 5 Å, so one would not expect any significant effects except in ultrathin layers. Accordingly, all previous studies[1–5] of the effect of a field have used YBaCuO films thinner than 100 Å. On the other hand, several studies (see Ref. 6 and the papers cited there) provide evidence that the field effect may be important even in bulk ceramic samples of high-T_c superconductors.

In this letter we are reporting a study of the effect of the spontaneous polarization of a ferroelectric on the electrical properties of relatively thick YBaCuO films (with thicknesses of about 1000 Å) in superconductor-ferroelectric-metal structures.

Films with a thickness of about 1000 Å were deposited on ferroelectric substrates, specifically, on the (001) plane of $BaTiO_3$ and on the plane of $LiNbO_3$ perpendicular to the polar axis. The substrates were 200–500 μm thick. The deposition was carried out by magnetron sputtering in an 9:1 argon–oxygen gas mixture at a total pressure of 120 mtorr in the chamber. The substrate temperature was about 700 °C. As usual, the films were cooled to 450 °C and held in an oxygen atmosphere at a pressure of 1 bar for 15 min in order to produce the orthorhombic phase of the film. The c axis was oriented perpendicular to the plane of the substrate. Silver was deposited (as a gate) on the back of the substrate, and indium contacts were formed on the film surface for resistance measurements by the van der Pauw method. Figure 1 shows a schematic diagram of the structure. The temperature dependence of the resistance was measured at currents of 10–50 μA. The measurement current in the mixed resistive state was a few tens of milliamperes; the magnetic field was applied in the direction perpendicular to the plane of the film. The leakage current did not exceed 10 mA at voltages of 3–4 kV applied to the structure.

Figure 1 shows the temperature dependence of the resistance of films on substrates of single-domain $LiNbO_3$ crystals, on planes perpendicular to the direction of the spontaneous polarization. The $+P$ and $-P$ represent films on substrates with polarizations directed toward the film and away from the film, respectively. The films were deposited in a common process under identical conditions.

As can be seen from Fig. 1, the temperatures at which the transition is completed ($R=0$) in the films on the substrates in the $+P$ and $-P$ states differ by almost 40 K, while the transition onset temperatures are nearly the same. The critical current densities at 4.2 K are also quite different: $j_{-P}/j_{+P}\approx 3$.

Unfortunately, the coercive field in bulk lithium niobate single crystals at temperatures far from the phase-transition point (≈ 1200 °C) is so high that polarization switching is essentially impossible. From this standpoint, barium titanate, $BaTiO_3$, is more convenient; in this case the polarization can easily be switched in fields on the order of 10^3 V/cm.

Figure 2 shows the temperature dependence of the resistance of a YBaCuO film on a $BaTiO_3$ substrate as the polarization is switched by an external electric field. Here, in contrast with Fig. 1, we see basically a shift of the $R(T)$ dependence, and the magnitude of the shift is much smaller, no greater than 1 K.

Figure 3 shows the change in the resistance of a film on a $BaTiO_3$ substrate in the mixed resistive state (77 K) as the substrate polarization is switched from the $+P$ state to the $-P$ state by an external electric field. Also shown here is a ferroelectric hysteresis loop, measured by the Sawyer–Tower method in the same substrate. Figure 3 reveals a clear correlation between the switching of the polarization and the change in the resistance.

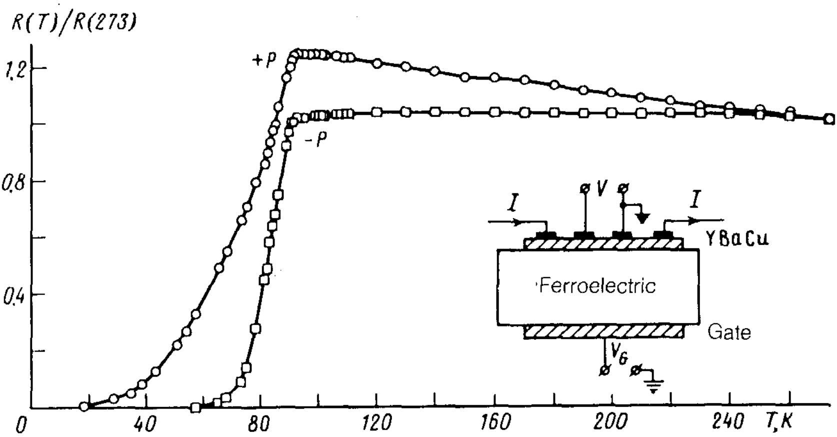

FIG. 1. Temperature dependence of the resistance of films on $LiNbO_3$ substrates polarized in opposite directions. The polarization was directed either toward the film ($+P$) or toward the gate ($-P$).

Let us discuss the results briefly. The effect of the polarization is greater in the case of lithium niobate: The shift of the temperature at which the transition is completed is nearly 40 K. This is the largest shift which has ever been observed experimentally. The spontaneous polarization (i.e., the surface charge density) in lithium niobate is $\simeq 70\ \mu C/cm^2$. Let us compare these results with experimental results on the field effect in (high-T_c superconductor)-insulator-metal structures. In Ref. 4, for example, a $SrTiO_3$ film with $\epsilon = 40$ was used as the insulator, and the external electric field reached 6×10^6 V/cm. The surface charge density is then $\epsilon\epsilon_0 E \simeq 20\ \mu C/cm^2$, i.e., smaller by a factor of several units than for the spontaneously polarized $LiNbO_3$. Correspondingly, the polarization in lithium niobate should give rise to effects greater than the field effect in Ref. 4, and this is just what we see experimentally. In the case of barium titanate the component of the spontaneous polarization normal to the surface is about 7 $\mu C/cm^2$ in our crystals (at 77 K, $BaTiO_3$ is in a trigonal phase with a $\langle 111 \rangle$ polar axis). This circumstance should give rise to effects an order of magnitude smaller than in the case of lithium niobate, and, again, this is roughly what we see experimentally. We should also mention here that the observed effect in the case of $LiNbO_3$ may be due to not only the direct influence of the polarization on the film resistance but also the influence of the polarization on the deposition process itself.

In summary, this study has shown that the spontaneous polarization of ferroelectrics leads to changes in the properties of high-T_c superconducting films which are qualitatively the same as the changes caused by a field in (high-T_c superconductor)-insulator-metal structures. Quantitatively, the effect of the spontaneous polarization can be even greater.

The strong effects in relatively thick films ($\simeq 1000$ Å) at a Thomas–Fermi screening length $\lesssim 5$ Å show that the physics of these observed effects may involve pinning processes in the course of which the electric field (the polarization) changes the pinning force at the film surface and thus affects the lattice of vortices penetrating completely through the film. A final resolution of the nature of the observed effects will of course require further research.

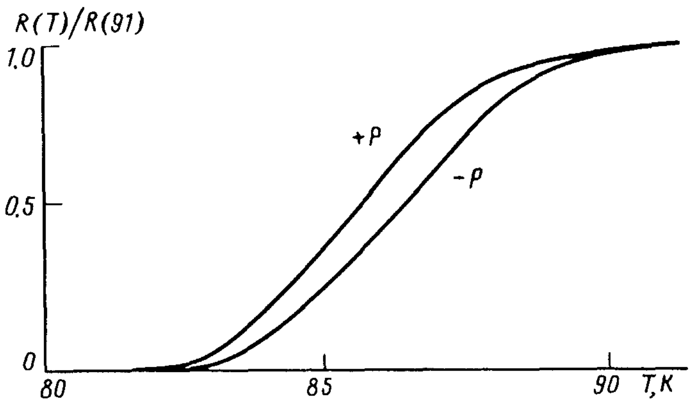

FIG. 2. Temperature dependence of the resistance of a film on a $BaTiO_3$ substrate as the substrate polarization is switched by an electric field of 10 kV/cm.

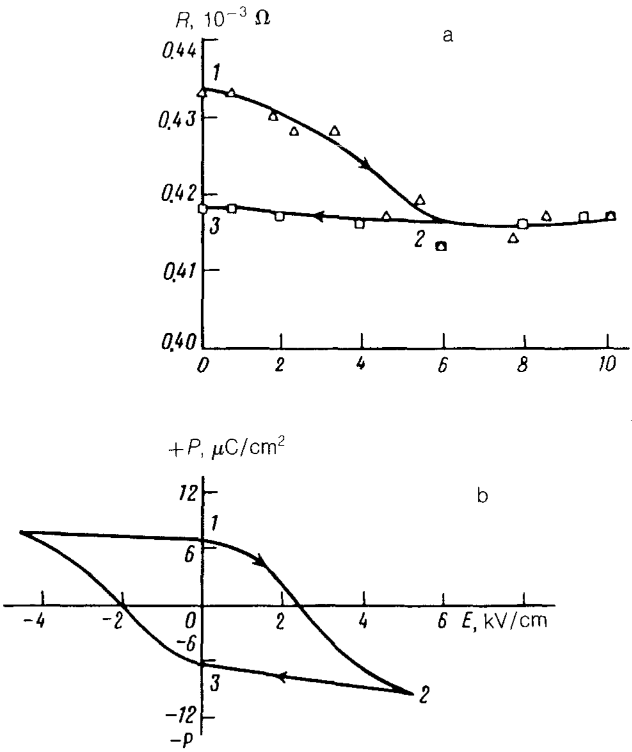

FIG. 3. a—Change in the resistance of a film on a $BaTiO_3$ substrate as the polarization of the substrate is switched; the film is in the mixed resistive state $9T = 77$ K, $H = 1.2$ kOe. b—Ferroelectric hysteresis loop in the same structure at 77 K.

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