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The influence of ferroelectric polarization on the superconductivity in ultrathin high- T_c films

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Abstract

Ferroelectric–superconductor heterostructures consisting of GdBa₂Cu₃O_{7-x} and YBa₂Cu₃O_{7- δ} channels and Pb(Zr_xTi_{1-x})O₃ (PZT) gate insulators were studied theoretically. We have shown that the remnant polarization P_r of the PZT gate induces a change in the carrier concentration n_h of the superconductor, supposing direct action of the Coulomb field on the carriers. The dependences of the superconducting transition temperature T_c on n_h were calculated in two cases: in the two-band model and in the phenomenological model with a parabolic dependence of T_c on n_h . The ferroelectric field effects are maximum in the superconducting film sample at a doping level where the transition temperature T_c as a function of n_h has the largest slope dT_c/dn_h . In agreement with the experiment, the shifts of T_c in the ferroelectric field $|\Delta T_c(\pm P_r)|$ decrease when the film thickness d increases. The form of the calculated $T_c(V_g)$ dependences on the gate voltage V_g can be related to the ferroelectric hysteresis loop of the PZT gate insulator.

1. Introduction

In recent years many efforts have been made to create novel high- T_c superconducting threeterminal devices such as flux-flow transistors [1] and field-effect transistors [2]. The silicon field-effect transistor has become the cornerstone of the modern semiconductor industry and technology. Ongoing efforts have been made to modulate superconductivity in high- T_c films with an applied static electric field [3–6]. However, these modulations have been observed in ultrathin YBa₂Cu₃O_{7- δ} (YBCO) films with thickness of 1–10 nm [7]. Device performance might be improved by using a relaxor ferroelectric such as Pb(Zr_xTi_{1-x})O₃ (PZT) as a gate insulator. Recently, ferroelectric field-induced effects have been observed in PZT/GdBa₂Cu₃O_{7-x} (GBCO) and PZT/YBCO heterostructures [8, 9]. Previously, in [10], qualitative estimations of the influence of the spontaneous polarization on T_c in ferroelectricsuperconductor structures were made. Relatively large ferroelectric field effects in high- T_c superconducting films were predicted. The transverse static electric field effects in high- T_c cuprates were studied theoretically in [11]. Using the technique of gate-induced doping in a field-effect transistor configuration, superconductivity at 52 K was induced in C₆₀ [12]. In [13] a new class of superconductors: insulating molecular crystals that are made metallic through charge injection, was presented. The goal in this article is to study the ferroelectric field effects in PZT/GBCO and PZT/YBCO heterostructures theoretically. We show that the remnant polarization \vec{P}_r of PZT changes the carrier concentration n_h of the superconducting films, using a mechanism of direct action of the Coulomb field on the carriers. We use the two-band model [11, 14] and the phenomenological model [15] for the calculation of the dependences of T_c in GBCO and YBCO on the carrier concentration. The ferroelectric field effects themselves are explained in terms of a phase diagram ($T_c(n_h)$).

2. Calculation of the dependence of T_c on the carrier concentration in YBa₂Cu₃O_{7- δ} in the two-band model

The superconducting transition temperature in the interband model [11, 14, 16] can be obtained from the following system of equations (for details of the consideration of the intraband Coulomb and electron–phonon interactions, see [16]):

$$\Delta_{1} = W \rho_{2} \Delta_{2} \int_{E_{0}-\zeta}^{E_{c}-\zeta} \frac{\mathrm{d}E}{\sqrt{E^{2} + \Delta_{2}^{2}}} \tanh \frac{\sqrt{E^{2} + \Delta_{2}^{2}}}{2k_{B}T}$$

$$\Delta_{2} = W \rho_{1} \Delta_{1} \int_{-\zeta}^{E_{c}-\zeta} \frac{\mathrm{d}E}{\sqrt{E^{2} + \Delta_{1}^{2}}} \tanh \frac{\sqrt{E^{2} + \Delta_{1}^{2}}}{2k_{B}T}.$$
(1)

The equation

$$\frac{1}{4}W^2\rho_1\rho_2\int_{-\zeta}^{E_c-\zeta}\frac{\mathrm{d}E}{E}\tanh\frac{E}{2k_BT_c}\int_{E_0-\zeta}^{E_c-\zeta}\frac{\mathrm{d}E}{E}\tanh\frac{E}{2k_BT_c} = 1$$
(2)

corresponding to the gaps $\Delta_{1,2}(T_c) = 0$ determines the superconducting transition temperature. The constant of the interband interaction $W = U + V_{12}$, where U is the interband Coulomb interaction and V_{12} is the corresponding contribution of the electron-phonon interaction. The pair-transfer interband interaction constants, $W_{\sigma_1\sigma_2\sigma_3\sigma_4}$, with $\sigma_1 = \sigma_2 = \sigma$ and $\sigma_3 = \sigma_4 = \sigma'$, where σ_n are the band indices and $\sigma \neq \sigma'$, are used [11]; i.e. in our model $W_{1122} = W_{12} = W$. In equations (1) and (2) the densities of states of the quasi-two-dimensional broad band and narrow band per spin and unit cell for the parabolic spectra are denoted by $\rho_{1,2}$. The top and bottom energies of the narrow band are $-E_0$ and $-E_c$; $\zeta = -\mu$ is the chemical potential of the holes.

From our model it follows that the dependence of T_c on the hole concentration n_h is determined by the chemical potential $\zeta(n_h)$. For the calculation of the doping dependence of ζ the equation

$$\rho_1 \int_0^{E_1} f(E) \, \mathrm{d}E + \rho_2 \int_{E_0}^{E_c} f(E) \, \mathrm{d}E = p \tag{3}$$

is used. Here $f(E) = \{\exp[(E - \zeta)/k_BT] + 1\}^{-1}$, p is the number of holes per cell, and E_1 is the width of the broad band. We introduce the carrier concentration $n_h = p/c$, where c is the number of CuO₂ planes per cell. In agreement with the experimental results [17] for La_{2-x}Sr_xCuO₄, in this paper the picture where the electronic chemical potential μ is shifted

from the top of the valence band with n_h is used. From equation (3) it follows that

$$\zeta = \begin{cases} n_h / \rho_1 & \text{if } E_0 - \zeta \gg k_B T \\ (\rho_1 + \rho_2)^{-1} (n_h + \rho_2 E_0) & \text{if } \zeta - E_0 \gg k_B T \text{ and } E_c - \zeta \gg k_B T \\ \frac{n_h}{\rho_1} + \frac{1}{2} E_1 - \frac{\rho_2}{\rho_1} k_B T \ln 2 & \text{if } \zeta = E_0. \end{cases}$$

For the YBa₂Cu₃O_{7- δ} superconductor we use the parameter values $\rho_1 = 0.25 \ 1 \ eV^{-1}$ cell, $\rho_2 = 1.5 \ eV^{-1}$ cell, $W = 0.5 \ eV$, $E_c = 0.92 \ eV$, $E_1 = 2.7 \ eV$, and $E_0 = 0.68 \ eV$. From our calculations it follows that one band is narrow. The width of this band is determined by the difference $E_c - E_0 = 0.24 \ eV$. The first set of parameters differs from that used in [11] because in [11] $n_h = (p - p_0)/c$, where $p_0 = \rho_1(E_1/2)$ (in this work $p_0 = 0$). The calculated dependence of T_c on $\Delta p \equiv n_h$ for YBCO in comparison with the experimental data [18] is depicted in figure 1. The agreement is good, as in the case $p_0 \neq 0$ [11]. We found that the static electric field effects [11] do not themselves depend on the difference of these two sets of parameters (obtained for the cases $p_0 = 0$ and $p_0 \neq 0$). The shift of the chemical potential with the carrier concentration changes the phase volume for pair-transfer scattering of electrons between the bands and leads to the observed bell-like dependence of T_c on n_h . For GdBa₂Cu₃O_{7-x} we use the same phase diagram as for YBa₂Cu₂O_{7- δ} [19].



Figure 1. The theoretical dependence of T_c on the hole concentration Δp for YBCO (solid line) in comparison with experiment (circles) [18].

We use parabolic electronic bands in this work (consideration of a hyperbolic band structure is also possible). However, in doing so we add nothing to the calculation of the ferroelectric field effects themselves, because in the case of the parabolic bands the experimental dependence of T_c on the carrier concentration is reproduced well. We emphasize that our theory of ferroelectric field effects does not depend on a special microscopic model of the superconductivity. For our purposes the model of high- T_c superconductivity in the Van Hove scenario is appropriate [20–24].

3. Ferroelectric field effects in superconducting films

A diagram showing the interface geometry with the direction of the electric fields is presented in figure 2. Electrostatic screening, as described in the Thomas–Fermi model, counteracts



Figure 2. A diagram showing the interface geometry with the direction of the electronic fields; Ω is a current source.

the penetration of the ferroelectric field into a superconducting film and, thus, reduces the ferroelectric field effects. The Thomas–Fermi charge screening length equals [11]

$$l_{TF} = \left(\frac{\hbar^2 \pi \epsilon_s}{4m_e k_F e^2}\right)^{1/2} = \left(\frac{\epsilon_s E_{F_0}}{4\pi^2 e^2 n_0}\right)^{1/2} \tag{4}$$

where ϵ_s is the dielectric constant of the superconductor; E_{F_0} and n_0 are the Fermi energy and the carrier concentration in the sample in the absence of the ferroelectric field (the remnant polarization $P_r = 0$), respectively, and e is the electron charge. For YBCO, the carrier concentration $n_0 \sim 5 \times 10^{21}$ cm⁻³, $\epsilon_s = 26$, and thus $l_{TF} = 0.5$ nm. In [25, 26], the estimates $l_{TF} = 0.5$ -1 nm have been obtained. In high- T_c superconductors the low density of charge carriers leads to a relatively large penetration depth. If a transverse ferroelectric field penetrates into a superconductor, its response to the field will depend sensitively on the value of the parameter $\tau = l_{TF}/\xi_z$, where ξ_z is the coherence length in the ferroelectric polarization direction. In analogy with the electrostatic field effects [11], in our model the effects of the ferroelectric field are connected with the changes of the carrier concentration in the remnant polarization field and, as a consequence, with the change of the chemical potential $\zeta(P_r)$.

In the framework of the Thomas–Fermi approximation, we obtained for the carrier concentration of the superconductor $n_s(z)$ in the remnant polarization field P_r (the coordinate $\vec{z} \parallel \vec{P}_r$)

$$n_s(z) = \frac{P_r}{el_{TF}} \exp(-z/l_{TF}).$$
(5)

Next, we also consider the case where the total charge induced by the ferroelectric field participates in the conductivity, and the surface (σ_s) and bulk (σ_b) conductivities are equal. This assumption is more justified in the case $\tau = l_{TF}/\xi_z \ge 1$. Using equation (5), the ferroelectric polarization-induced changes of the carrier concentration Δn averaged over the thickness *d* of a superconducting film are given by the expression

$$\Delta n = \frac{1}{d} \int_0^d n_s(z) \, \mathrm{d}z = \frac{P_r}{ed} (1 - \exp(-d/l_{TF})). \tag{6}$$

It is clear that for $d \gg l_{TF}$, $\Delta n = \Delta Q/d$, where $\Delta Q = e^{-1}P_r$ is the ferroelectric fieldinduced surface carrier density (for the surface layer). If the surface conductivity σ_s and the bulk conductivity σ_b are not equal,

$$\Delta n = \frac{P_r}{ed} \frac{\sigma_s}{\sigma_b} (1 - \exp(-d/l_{TF}))$$

which is a modification of equation (6).

The expression for the chemical potential $\zeta(P_r)$ follows from equation (3) for ζ on making the replacement $n_h \rightarrow n_h + n_{P_r}$ with $n_{P_r} = v \Delta n$, where Δn is determined by equation (6). Thus, the ferroelectric field changes the average carrier concentration n_h per CuO₂ plane (ferroelectric field-induced doping). The thickness of superconducting films fundamentally influences n_{P_n} and the magnitude of the ferroelectric field-induced effects.

Using equation (2), we obtain the shift for the superconducting transition temperature of films in a ferroelectric polarization field defined as $\Delta T_c(P_r) = T_c(P_r) - T_c(0)$. In the positive ferroelectric polarization field P_r , the change of the number of holes (electrons) per cell $n_{P_r} < 0$ and, if $(dT_c/dn_h) > 0$ (see figure 1), it can be shown that $\Delta T_c(P_r) < 0$. For the negative polarization P_r , $n_{P_r} > 0$ and $\Delta T_c(-P_r) > 0$. In the case $(dT_c/dn_h) < 0$, these shifts are opposite. This behaviour correlates with experiment [8, 9], which shows that the ferroelectric field effects are really the bulk effects.

4. Calculations for Pb(Zr_xTi_{1-x})O₃/GdBa₂Cu₃O_{7-x} and Pb(Zr_xTi_{1-x})O₃/YBa₂Cu₃O_{7- δ} heterostructures

Next, on the basis of the expression for n_{P_r} , equations (2), (5), (6), and the modification of equation (6), we calculate the dependences of the shifts $\Delta T_c(P_r)$ in Pb(Zr_xTi_{1-x})O₃–GBCO heterostructure. For this structure the ferroelectric field effect in ultrathin films of GBCO was measured by Ahn *et al* [8]. In the samples studied in [8] the remnant polarization P_r of the ferroelectric relaxor PZT equals 10 μ C cm⁻² and the coercive field is 100 kV cm⁻¹. In the external electric field \vec{E} in equations (5) and (6), the substitution $P_r \rightarrow P(E) = P(V_g)$ (V_g is the gate voltage) should be made. Shifts of the superconducting transition temperature of 7 K were measured for two different PZT/GBCO/PBCO (PrBa₂Cu₃O₇) heterostructures for both polarization states ($\pm P_r$) of the PZT film [8]. In [8], during the fabrication of the heterostructures, the ferroelectric film was uniformly poled. As a result, it was not possible to measure the resistivity for the unpolarized (randomly polarized) state of a ferroelectric. In [8] for GBCO the ferroelectric polarization-induced shift ΔT_c ($\pm 10 \ \mu C \ cm^{-2}$) was measured, according to our calculations, for $T_c(0) = 34.5$ K. Using equation (5) and equations (2), (3) and supposing z = d, i.e.

$$n_s(d) = \frac{P_r}{el_{TF}} \exp(-d/l_{TF})$$

we calculated $\Delta T_c = T_c(+10 \ \mu \text{C cm}^{-2}) - T_c(-10 \ \mu \text{C cm}^{-2}) = 7 \text{ K}$ for d = 2 nm for GBCO using the two-band model with $l_{TF} = 0.35 \text{ nm}$ and the phenomenological parabolic dependence $T_c(n_h)/T_{c,max} = 1 - 82.6(\Delta p - 0.16)^2$ [15] with $l_{TF} = 0.54 \text{ nm}$, respectively. The values of $l_{TF} = 0.35 \text{ nm}$ and 0.54 nm coincide approximately with the value $l_{TF} = 0.5 \text{ nm}$ for YBCO [25]. Other fitting parameters are used to fit the curve $T_c(n_h)$ in the two-band model. The ferroelectric field effects themselves are practically independent of the two-band model (see below). Then we calculated the dependences of ΔT_c on film thickness *d* for GBCO film and the remnant polarization $P_r = \pm 10 \ \mu \text{C cm}^{-2}$. These curves are presented in figure 3. As is seen from figure 3, the shifts ΔT_c with *d* are large at d = 1.2 nm, i.e. in the film sample of GBCO with a thickness of one cell unit, and ΔT_c is practically zero at d = 4 nm.

We also use another picture, namely the modified equation (6) with $\beta = \sigma_s / \sigma_b$, for the calculation of ΔT_c . For GBCO, we have determined from the experimental data [8]



Figure 3. The dependences of $\Delta T_c = T_c(+10 \ \mu \text{C cm}^{-2}) - T_c(-10 \ \mu \text{C cm}^{-2})$ for the GBCO film on the thickness *d* in the two-band model for $l_{TF} = 0.35$ nm (chain line) and $\beta = 0.02$ (dashed line); and in the case of the phenomenological parabolic dependence $T_c(\Delta p)$ [15] for $l_{TF} = 0.54$ nm (solid line A) and $\beta = 0.1$ (solid line B). The open square corresponds to the experiment [8].

 $\beta = 0.02$ and $\beta = 0.1$ using the two-band model and the phenomenological parabolic dependence [15] of $T_c(n_h)$, respectively. A shift Δn_{P_r} directly changes the carrier concentration Δp in the formula for $T_c(n_h)/T_{c,max}$. We have calculated the dependence $\Delta T_c(d)$ with these β -values (figure 3). As is seen from figure 3, the shifts ΔT_c do not equal zero up to d = 20 nm. These values of β are close to those obtained in [9, 11]. The ferroelectric remnant polarization-induced shifts at $P_r = \pm 10 \ \mu \text{C cm}^{-2}$ in figure 3 change as d^{-1} . A tendency towards such a dependence on d^{-1} for YBCO films was observed in [9]. In [11], $\beta = \sigma_s/\sigma_b$ also depends on d.

In our opinion the first picture (with l_{TF} , equation (5)) is more appropriate for thin inhomogeneous films samples, and the second picture (with β) is more appropriate for relatively homogeneous films.

The shifts $\Delta T_c(P_r)$ strongly depend on P_r . For positive ferroelectric remnant polarization and $(dT_c/dn_h) > 0$, $\Delta T_c(\vec{P}_r) < 0$ in agreement with the experimental results [9]. For negative polarization, $-\vec{P}_r$, the shifts $\Delta T_c(-\vec{P}_r) > 0$ are also in agreement with experiment [9].

We have also calculated the dependences of $\Delta T_c(\pm P_r)$ on the remnant polarization P_r and the film thickness *d* in YBCO. As follows from our calculations, the ferroelectric field effects are similar in the two compounds.

The ferroelectric field effect in ferroelectric–superconductor film (YBCO sample) structures was studied experimentally in [9, 27]. This structure [9] consists of an YBa₂Cu₃O_{7- δ} channel and a Pb(Zr_{0.54}Ti_{0.46})O₃ gate insulator. In order to prevent the degradation of the YBCO film, a thin SrTiO₃ buffer layer was inserted between the YBCO and PZT in [9]. The PZT films were epitaxially grown in [9] and they revealed high remnant ferroelectric polarizations of up to 61 μ C cm⁻². Charging effects induced by the polarization P_r were observed in a YBCO film thinner than 20 nm. The reversion of the ferroelectric polarization state leads to a nonvolatile shift of T_c in YBCO.

Using the experimental [9] values of

$$P_r(V_g = 0) - (-P_r(V_g = 0)) = \Delta P_r = 45 \ \mu \text{C cm}^{-2}$$



Figure 4. The dependences of $\Delta T_c = T_c (22.4 \ \mu \text{C cm}^{-2}) - T_c (-22.6 \ \mu \text{C cm}^{-2})$ on the film thickness *d* for YBCO calculated using the two-band model and $\beta = 0.0035$ (dashed line) and in the case of the parabolic dependence of $T_c (\Delta p)$ [15] and $\beta = 0.017$ (solid line) in comparison with the experiment [9] (solid squares).

and $\Delta T_c = 0.85$ K, we have calculated from modified equation (6) for a 8.8 nm thick YBCO film [9] and the parabolic dependence $T_c(n_h)$ [15] the value $\beta = 0.0035$. This value of β is an order of magnitude smaller than β_e estimated from the equation $\Delta T_c/T_c = \beta_e \Delta n_h/n_h$. In [9], β_e has no physical meaning. We obtained $T_c(0) = 35.3$ K. The dependence of ΔT_c for YBCO on the film thickness *d* was calculated using equations (5), (6) ($\beta = 0.0035$) and the parabolic dependence of $T_c(n_h)$ ($\beta = 0.017$) (figure 4). In [9], for YBCO films thinner than 7 nm no superconductivity was found at all. But in YBa₂Cu₃O_{7- δ}/PrBa₂Cu₃O₇ superlattices [28], superconductivity was observed in the YBCO film with a thickness of up to one cell unit. We have calculated the dependence $\Delta T_c(d)$ up to 1.2 nm. For the thicknesses smaller than d = 4 nm in [25], $T_c(0)$ depends on d, but we have not taken this dependence into account. In general, for d > 7 nm this dependence is inessential and at d < 4 nm lowers $|\Delta T_c|$. The calculated $\Delta T_c(d)$ has the d^{-1} -behaviour exhibited in the experiment [9].

Figure 5 shows the dependence $T_c(V_g)$ in a YBCO film with the thickness d = 8.8 nm for positive and negative gate voltages V_g . We have used the polarizations $P(V_g)$ of the 20 nm SrTiO₃/1000 nm PZT gate insulator of a YBCO/PZT/Au structure with a SrTiO₃ buffer layer measured at 100 K ([9]) for the calculation of $T_c[P(V_g)]$. Our calculated $T_c[P(V_g)]$ are, in general, consistent with the experiment [9] and with the ferroelectric charging effect. The changes of the critical temperature T_c with V_g clearly reflect the ferroelectric hysteresis loop of the PZT gate insulator.

The relatively small values of $\beta = \sigma_s/\sigma_b$ are connected in YBCO as in GBCO with the circumstance that the surface conductivity σ_s is substantially smaller than the bulk conductivity σ_b (cf. [11]), which indicates the presence of interface traps, defects, and an ionic conduction. It was also pointed out from theoretical considerations [29] using the Ginzburg–Landau equation for the order parameter Ψ that a small oxygen nonstoichiometry or disorder in the CuO₂ planes near the channel/insulator interface suppresses the modulation of T_c . Small values of β have been reported for the charging effects in nonferroelectric–superconductor field-effect transistors [30].



Figure 5. Superconducting transition temperature T_c for the 8.8 nm YBCO film versus gate voltage (the corresponding $P(V_g)$ in PZT). Solid squares with chain lines are experimental dependences [9] and the dashed line and solid line (which practically coincide) are the theoretical curves in the two-band model ($\beta = 0.0035$) and in the case of the phenomenological parabolic dependence of $T_c(\Delta p)$ ($\beta = 0.017$), respectively.

5. Conclusions

In this work the ferroelectric field effects in ultrathin cuprate films have been investigated in ferroelectric-superconductor structures. The dependence of the superconducting transition temperature T_c on the carrier concentration n_h at the remnant polarization $P_r = 0$ is calculated for YBa₂Cu₃O_{7- δ} in the two-band model with improved parameters, and in the picture where the chemical potential μ is shifted from the top of the wide valence band with doping. The use of fitting parameters, which all have clear physical meaning, allows us to achieve a good agreement of the dependence $T_c(n_h)$ with the experiment for YBCO. Dependences of the T_c for ultrathin films of GdBa₂Cu₃O_{7-x} and YBa₂Cu₃O_{7- δ} on the ferroelectric remnant polarization P_r of the relaxor Pb(Zr_xTi_{1-x})O₃ have been calculated using the two-band model [11, 14, 16] and the phenomenological parabolic dependence $T_c(n_h)$, respectively. We suppose a direct charge-transfer mechanism induced by the ferroelectric polarization $P(V_g)$. The ferroelectric field-induced shifts of T_c depend on the strength and polarity of the P_r . For the maximum ferroelectric field effect the high- T_c superconducting film has to be away from optimal doping and has to be closer to the maximum of the slope dT_c/dn_h . In agreement with experiments, the shifts $|\Delta T_c(\pm P_r)|$ decrease when the film thickness d increases. The theory agrees qualitatively and, in some cases, quantitatively with the experiment for ultrathin GdBa₂Cu₃O_{7-x} and YBa₂Cu₃O_{7- δ} films. As follows from our analysis, in the first case the number of fitting parameters of the theory is equal to 1 (the ratio $\beta = \sigma_s / \sigma_b$) and in the second case the fitting parameter was the Thomas–Fermi screening length $l_{TF} = 0.35-0.5$ nm for GdBa₂Cu₃O_{7-x} (from estimations, $l_{TF} = 0.5$ nm follows [25]). The other fitting parameters are used to fit the dependence $T_c(n_h)$.

The microscopic theory of superconductivity in high- T_c systems is a debatable topic. The two-band model [11, 14, 16] for cuprates has helped to explain some experimental data. The ferroelectric polarization effects do not depend themselves on the two-band model and are explained in terms of a known phase diagram of cuprates where T_c changes systematically with the carrier concentration.

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