



Study of SrTiO₃ thin films grown by sputtering technique for tunnel barriers in quasiparticle injection contacts

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ABSTRACT

High-quality, *c*-axis oriented YBa₂Cu₃O_{7-x}/SrTiO₃/Au (YBCO/STO/Au) planar structures were fabricated *in situ* by direct current/radiofrequency inverted-cylinder magnetron sputtering on (001) STO oriented substrates. The sandwich-type structures were patterned to transistor dimensions by standard ultraviolet-photolithography and Ar etching. The current transport mechanism in the very thin STO barriers (2–30 nm) was examined by measuring the tunneling *G* as function of temperature (*T*), and bias voltage (*V*). It was found that resonant tunneling and hopping via a small number of localized states (LS) are responsible for electronic conduction in the insulating material. Elastic tunneling was observed for the case of a nominal 2 nm thick STO-barrier with an energy gap $\Delta \approx 20$ meV in the (001) direction of YBCO. On the other hand, inelastic hopping transport via *n*-LS dominated for STO barrier thickness $d > 2$ nm. *G* of the lowest-order hopping channel (hopping via two LS) exhibits the characteristic *T* and *V* dependences: $G_2^{\text{hop}}(T) \propto T^{4/3}$, $G_2^{\text{hop}}(V) \propto V^{4/3}$, respectively. Increasing the thickness of the STO barriers, hopping channels of higher order contribute more and more to the current transport as proven by measuring the *T* and *V* dependences. A crossover to variable range hopping behavior has been observed for junctions with thicker barriers ($d \geq 20$ nm) in the high-*V* or high-*T* regime. By fitting the experimental data to theoretical models, physical parameters of the LS could be determined. For instance, the value of the localization length or radius of the localized state was determined to be $\sim 4.6 \times 10^{-8}$ cm which corresponds to the lattice constant of the STO unit cell. A value of $\sim 6 \times 10^{19}$ (eV)⁻¹ cm⁻³ was calculated for the density of LS and the average barrier height was estimated as ~ 0.4 eV.

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1. Introduction

The perovskite-type STO has been frequently used as a somewhat ideal material for the growth of YBCO thin films. This is due to its good chemical stability (no reaction with YBCO), good lattice matching (the STO cubic lattice parameter, *a*, (0.3905 nm) is close to the lattice parameters *a* and *b* of the YBCO unit cell) and the good thermal expansion coefficient matching. In addition, STO thin films show very good insulating properties, with a breakdown *V* as high as 10⁵ V/cm. Such feature makes this material suitable as artificial barrier for high-*T* superconductor quasiparticles injection devices as well as for capacitive charging in superconducting field effect transistors [1]. In addition, the non liner dependence of the dielectric constant (ϵ), on the electric field (*E*), is important for the development of tunable microwave devices [2].

Up to now, the mechanism of current transportation across STO-thin films has been investigated for *c*-axis oriented Au/STO/YBCO and YBCO/STO/YBCO junctions [3] with a STO barrier layer ranging from 200 to 500 nm. Analysis of the data showed that the behavior of the junctions could be well described within the framework of Mott's

variable range hopping (VRH) via LS theory. This theory predicts that the typical length of a hop, the variable range hopping length (l_{VRH}), increases with decreasing *T* as $T^{-1/4}$. This *T* dependence describes the electronic transport in a variety of insulating and semiconducting materials over a wide range of *T*.

The electron wave function within a localized state decays exponentially over a distance known as the localization length (α^{-1}). This physical quantity specifies the importance of the presence of LS in the barrier and consequently the dominant conduction process at a determined *T*. In case of a thin layer with a thickness comparable to α^{-1} , it is not expected that the existence of LS alters the conduction process significantly [4]. Direct tunneling between the two electrodes is then the dominant process in this film. Nevertheless, as *d* is increased *G* shows a more complex dependence on *T* and *V*.

A very strong influence of the LS on *G* is expected when *d* is much greater than α^{-1} but smaller than l_{VRH} . In this range the elastic and the inelastic tunneling process via a small number of LS exhibits individual transport characteristics — which are typical for such small number of LS participating in the conduction process — before the VRH-mechanism is set in for higher thickness, *T* or *V*.

The hopping transport via two LS, which forms an optimal conductive chain, has been well explained by Glazman and Matveev

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(GM) [5]. In case of tunneling through two LS ($n=2$) this microscopic model predicts a T and V dependence of the conductance (G), given by $G_2(T)=\alpha_2 T^{4/3}$ for $eV \ll k_B T$ (k_B is the Boltzmann constant), and $G_2(V)=\alpha_2 V^{4/3}$ for $eV \gg k_B T$, respectively. An increase of T , V or d favors hopping along chains containing more consecutive LS ($n>2$). This led to an increasingly non linear dependence of G on the T and/or V .

In this paper, the transport mechanism in c -axis oriented YBCO/STO/Au planar-type injection junction is analyzed using the GM theory. At first, the analysis of the normal current transport including the T dependence of G and the shape of the current–voltage (I – V) curve in the high V region is described. Then α^{-1} and the density of LS, g , are estimated. Special attention is also paid to the possibility of elastic tunneling of quasi-particles in very thin STO barriers layers.

2. Experiment

The YBCO/STO/Au multilayers were grown *in-situ* on STO (100) substrates using a magnetron sputtering system equipped with sintered cylindrical YBCO, STO and disc-shaped Au targets. The junction fabrication process started with the deposition of a base YBCO layer with a thickness ranging from 50 to 80 nm onto the STO (100) substrate. The deposition was carried out by dc-sputtering in a 50% containing Ar/O₂ atmosphere at a total pressure of 52 Pa and at a direct current (dc)-power of 45 W. The substrate was maintained at 810 °C during film deposition. Under these conditions the deposition rate amounted to 4 nm/min as it was determined by means of Rutherford backscattering spectrometry. This method of analysis permitted also the determination of the film composition. After the film deposition no other additional annealing treatment was employed other than the very slow cooling down to room T in an O₂ atmosphere of 8.0×10^4 Pa. The so fabricated YBCO layers showed very good morphological, structural and electrical properties. Atomic-force microscopy (AFM) studies confirmed that the root-mean-square surface roughness of a 50–80 nm thick YBCO film deposited on STO (100) substrates was less than 2 nm on $50 \times 50 \mu\text{m}$ AFM scans. To provide such smooth surfaces is essential for overlayer growth, e.g., a very thin artificial barrier like STO, which must cover uniformly the effective device-area. Furthermore, it has to be shorts- and pin-hole-free in order to insure electrical insulation between both bottom electrode (YBCO) and counter electrode (Au). From X-ray diffraction (XRD) measurements, it could be concluded that the films were c -axis oriented, with a , b planes parallel to the substrate. The c -axis length resulted to be 11.683 Å, which corresponds to fully oxygenated films [6]. The full width at half maximum in the ω scan at the (500) reflection was measured to be approximately 0.4° , confirming both the very good crystalline quality and the high degree of orientation of this film. The superconducting transition temperature (T_c) was about 90 K as determined by an inductive method with a transition width less than 0.5 K. In order to fabricate the multilayered heterostructure (YBCO/STO/Au), the base YBCO-layer was — after the deposition — slowly cooled down to 590 °C in the same sputtering atmosphere.

The STO insulator layers with a thickness varying from 2 to 30 nm were grown on smooth c -axis oriented YBCO layers. The deposition was carried out using a cylindrical magnetron sputtering system also — but in the rf mode — at a total pressure of 4.7 Pa with a 1:1 Ar/O₂ mixture and a substrate T of 590 °C. At a radiofrequency-sputtering power of 75 W the deposition rate of STO amounted to 1.7 nm/min. After deposition of the STO layer, the chamber was flooded with oxygen at a pressure of 8.0×10^4 Pa, and then the bilayer was cooled down to 450 °C and held in the oxygen atmosphere for 1 h. This annealing process ensures full oxygenation of the base YBCO-layer. Once the annealing process was completed the bilayer was cooled down to about 100 °C, then the counter Au-layer with about 50 nm in thickness was deposited at an Ar-pressure of 3.0×10^4 Pa. At a dc-sputtering power of 20 W the deposition rate was 100 nm/min. A light reduction of T_c in about 1 K without broadening of the transition width was observed after STO and Au deposition.

The junction area was defined and etched down to the bottom electrode using standard lithographic techniques and Ar ion milling. During the etching process the 500 V Ar ion beam was incident at 90° to the substrate surface in order to avoid ramp-like etched patterns, especially during the first photolithographic step. Four junctions with areas varying from 12×5 to $40 \times 5 \mu\text{m}^2$ were fabricated in one chip. Prior to the preparation of Au-contacts for the electrical characterization of the junction, which was carried out by a lift-off method with a photoresist stencil, an isolation layer between the bottom and top electrode was provided. For this purpose, polycrystalline PrBa₂Cu₃O₇ was deposited by rf-sputtering at a power of 100 W. For the electrical measurements of the junctions, the substrate with the four structured micro-bridge was glued on a chip-mount specially designed, allowing for different choices of the measurement-geometry: both for the STO barriers and the YBCO microbridges. Then, Al wire bonds to Au contact pads were prepared using ultrasonic-bonding. Finally the entire set was placed on an integrated circuit-socket fixed to the sample holder. Fig. 1 shows the schematic cross-sectional view of the investigated YBCO/STO/Au test junctions after the patterning process. The micro-bridge setup allows that the current is fed into the circuit via low-ohmic contact pads, which are far apart from the microbridge to assure that the associated Joule heating can be neglected.

Two terminal I – V measurements were made from 300 to 4.2 K with the junctions enclosed in a μ -metal cylinder. Measurements from room T to 4.2 K have been performed in He-vapor using a He-can. For measurements at 4.2 K the junctions were immersed in liquid He. I – V characteristics were measured by the application of a small oscillating current (1–10 Hz) and the detection of the resulting oscillating V over the sample employing a differential amplifier. This differential amplifier is an integrated component of a special current source [7] which was specifically constructed for electrical characterization of injection and Josephson contacts.

3. Results and discussion

3.1. Electronic transport in very thin STO barriers via elastic tunneling

Fig. 2 (a) shows the I – V -characteristic of an YBCO/STO/Au junction with a nominally 2-nm thick STO barrier layer measured at 77 K, showing clear evidence of superconductor–insulator–normal metal (S – I – M) single particle tunneling through a planar structure. The inset to Fig. 2 displays the dynamic G , dI/dV , versus V reflecting the shape of the superconducting density of states $[N(E)]$. This quantity may be calculated from tunnel data by normalizing the superconducting to the normal state by means of the relationship $N(E)=(dI/dV|_S)/(dI/dV|_N)$. The first noticeable feature on the inset to Fig. 2(a) is the lack of a zero-bias anomaly (G peak at zero bias), probably indicating in the YBCO c direction [8] as the nominal current orientation of the planar junction, but it would also be consistent with tunneling straight along the YBCO a or b axis [9]. Well defined maxima are observed at about ± 20 meV, indicating the presence of energy gap structures at $\pm \Delta$. Thermal smearing of the dI/dV

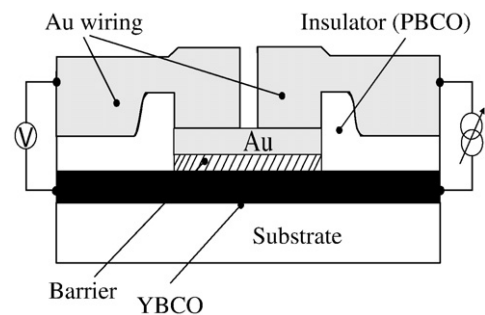


Fig. 1. Schematic cross sectional view of a tunnel junction with a YBCO/STO/Au layered structure.

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