Intrinsic Josephson properties in 
Pb$_{1-y}$Sr$_2$Y$_{1-x}$Ca$_x$Cu$_{2+y}$O$_{7+\delta}$ epitaxial films

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Abstract. We report the first observation of intrinsic Josephson junction (IJJ) characteristics in the Pb$_{1-y}$Sr$_2$Y$_{1-x}$Ca$_x$Cu$_{2+y}$O$_{7+\delta}$ (Pb1212) epitaxial film. Pb1212 epitaxial film has been grown on SrTiO$_3$ (100) substrates by a two-step growth technique. A small mesa structure ($S=4\mu$m$^2$) has been fabricated on a film surface using a standard photolithography and an Ar ion milling technique. The superconducting transition temperature of the IJJ is 43 K. At 4.2 K, the value of $J_c$ (the critical current density) is 2.2 kA/cm$^2$. The depth of the hysteresis defined as $\alpha=(J_r+J_i)/J_c$ is 0.89, where $J_r$ is the return current density. The temperature dependence of $J_c$ shows good agreement with Ambegaokar-Baratoff theory.

1. Introduction
A crystal of a high-$T_c$ cuprate is generally regarded as a stacked layer of the tunneling Josephson junctions. These junctions are called intrinsic Josephson junctions (IJJs). Since IJJs are based on the crystal structure itself and naturally formed, the surface of the junction is atomically flat and clean. For this reason, IJJs are investigated in various areas such as interlayer tunneling spectroscopy, macroscopic quantum tunneling, THz radiation, and so on. In ideal superconductor/insulator/superconductor IJJs, the current-voltage characteristics show a hysteresis and resistive branches which correspond to the resistive states of the individual IJJs. These characteristics are typically seen in IJJs of Bi$_2$Sr$_2$CaCu$_2$O$_8$, Bi$_2$Sr$_2$Ca$_2$Cu$_3$O$_{10}$, and Bi$_2$Sr$_2$CuO$_y$ which have crystal structures with a significantly large anisotropy around 100 or greater. Tl$_2$Ba$_2$CaCu$_2$O$_8$[1] and Tl$_2$Ba$_2$Ca$_2$Cu$_3$O$_{10}$[2] whose crystal structures are similar to those of Bi-family also exhibit IJJ characteristics. On the other hand, IJJ characteristics can be rarely observed in materials which have crystal structures with small anisotropy. In YBa$_2$Cu$_3$O$_7$, small hysteresis and resistive branches are observed only when the small junctions are fabricated in under-doped samples[3][4][5]. HgBa$_2$CaCu$_2$O$_{7+\delta}$[6] and RuSr$_2$GdCu$_2$O$_7[7]$ also exhibit characteristics similar to YBa$_2$Cu$_3$O$_7$. Recently, IJJ characteristics of Sm$_{2-x}$Ce$_x$CuO$_4[8]$, La$_{2-x}$Sr$_x$CuO$_4[9]$, Hg$_{1-x}$Re$_2$Ba$_2$Ca$_2$Cu$_3$O$_8[10]$, and Hg$_{1-x}$Re$_2$Ba$_2$Ca$_3$Cu$_4$O$_{10}[11]$ have also been observed. As far as our knowledge, these 12 types of cuprates have been reported to exhibit IJJ characteristics. We report the first observation of IJJ characteristics in Pb$_{1-y}$Sr$_2$Y$_{1-x}$Ca$_x$Cu$_{2+y}$O$_{7+\delta}$ (Pb1212). The crystal structure of Pb1212 is similar to that of YBa$_2$Cu$_3$O$_7$, HgBa$_2$CaCu$_2$O$_7$, and RuSr$_2$GdCu$_2$O$_7$. Due to the lack of single-crystal samples, the fundamental properties of Pb1212 have not been well investigated. We have obtained epitaxial films of Pb1212 by a two-step growth technique[12], and succeeded in the observation...
of IJJ characteristics.

2. Sample fabrication

Pb1212 epitaxial film was grown by a two-step technique consisting of a low temperature sputtering step and a high temperature ex situ growth step. For depositing the Pb1212 amorphous film on SrTiO₃ (100) substrate, we used the following sputtering conditions: composition of the sputtering target was (Pb₀.₇₅Bi₀.₂₅)₁ₛSr₂Y₀.₄₇₅Ca₀.₅₂₅Cu₂₀₂O₂, the sputtering gas pressure was 100 mTorr (60 sccm Ar and 15 sccm O₂), the anode voltage was 1.4 kV, and the substrate temperature was approximately 200 °C (the substrates were not heated). The deposition time was set to 90 min. The thickness of the Pb1212 thin film measured by a stylus-based profilometer was 2600 Å.

Pb1212 containers used for ex situ growth were made of polycrystalline pellets with the composition of PbSr₂Y₀.₃Ca₀.₇Cu₂₀₂O₂. For epitaxial growth, the amorphous film on SrTiO₃ substrate was placed in a pit formed at the center of one of the pellets. The other pellet was used as a lid for the growth container. The container containing the amorphous film was heated in a muffle furnace at 970 °C for 6 h under an O₂ atmosphere, then cooled to room temperature at a rate of 200 °C/h.

The results of out-of-plane x-ray diffraction measurements indicate the absence of impurity phase and complete c-axis alignment of the film. The results of in-plane x-ray diffraction measurements indicate that the film grow epitaxially on the substrate. The concentration of the film determined by energy dispersive x-ray spectroscopy was Pb₀.₇₀Sr₄(Y₀.₆₅Ca₀.₃₅)₁₄₇Cu₁₂₂₃O₂. In Pb1212, the Ca concentration x > 0.3 is necessary for the occurring of the superconducting transition[13], and approximately one-fourth of Pb ions in the block layer is substituted by Cu ions.

A subsequent quenching treatment is also necessary to make the film superconducting. In this treatment, the film was placed in a quartz tube, then heated at 815 °C in air for 1 h. Within two seconds after removing the quartz tube from the furnace, it was placed in liquid nitrogen. This procedure increases the hole concentration, decreases crystallographic disorder, and consequently results in the film being superconducting[13].

Ag (50 nm) was evaporated on a film surface as a contact electrode. Then, small mesa structures (S = 4μm²) were fabricated on a film surface using a standard photolithography and an Ar ion milling technique. The electrode configuration we employed is three-terminal type. The number of layered Josephson junctions, 21 layers, is controlled by the milling time. I – V characteristics of IJJs are observed by an analog oscilloscope.

3. Results and discussion

The superconducting transition temperature of the IJJ was 43 K. Hysteretic current-voltage characteristics and multiple branches, which are peculiar features of IJJs, were observed below 39 K. I – V characteristics of IJJs at 4.2 K are shown in Fig. 1. The value of Jc (the critical current density) is 2.2 kA/cm². The depth of the hysteresis defined as α = (Jc + Jr)/Jc is 0.89, where Jr is the return current density. The McCumber parameter (a measure of the damping of IJJs) is estimated to be 110 from the value of α. The fact that multiple branches can be observed near Tc and large values of α and βc suggest that the Pb1212 is a highly anisotropic cuprate like Bi-family. However, the ab-plane resistivity measurement in magnetic fields shows that the coherent lengths of Pb1212 are approximately ξab(0) = 2.5 nm and ξc(0) = 0.27 nm[13]. Therefore, the anisotropy, γ = ξab(0)/ξc(0) is 9.2, which is closer to that of YBa₂Cu₃O₇ (γ ~ 5) than that of Bi-family (γ ~ 100). The contradiction between characteristics of IJJs and the value of anisotropy may be peculiar to Pb1212 system. The gap voltage between the zero and the first branch is about 18 mV. This gap voltage decreases with increasing voltage or temperature.
The temperature dependence of the critical current of the first branch is shown in Fig. 2. The temperature and the critical current are normalized at $T_c$ and $I_c(4.2 \, \text{K})$. The solid curve represents the theoretical $I_c - T$ for a superconductor-insulator-superconductor (SIS) Josephson junction based on the Ambegaokar-Baratoff (A-B) formula\cite{14}

$$I_c(T) = \frac{\pi \Delta(T)}{2e R_N} \tanh \frac{\Delta(T)}{2k_B T}$$

where $R_N$ is the normal tunneling resistance, $2\Delta$ is the superconducting gap, and $k_B$ is the Boltzmann constant. Experimental result has a good agreement with A-B theory. This suggests that the IJJs of Pb1212 have a character of SIS-type Josephson junction. SIS characteristics have also been observed in IJJs of under-doped YBa$_2$Cu$_3$O$_7$\cite{5} and RuSr$_2$GdCu$_2$O$_7$\cite{7} whose crystal structures are similar to that of Pb1212.

4. Summary

Intrinsic Josephson characteristics in the Pb$_{1-y}$Sr$_2$Y$_{1-x}$Ca$_x$Cu$_{2+y}$O$_{7+y\delta}$ (Pb1212) epitaxial film have been observed. It is found that hysteretic current-voltage characteristics and multiple branches appears below 0.9 $T_c$. IJJs in Pb1212 shows under-damped behavior in spite of its small anisotropy. The temperature dependence of critical currents suggests that IJJs of Pb1212 have a character of SIS-type Josephson junction.

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