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Intrinsic tunneling spectroscopy for $Bi_{2-x}Pb_xSr_2CaCu_2O_{8+\delta}$ of nm-thickness mesa structure

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ABSTRACT

We have investigated intrinsic tunneling spectroscopy (ITS) with short-pulse bias to mesa structures consisting of several layers of intrinsic Josephson junction superlattices of $Bi_{1.8}Pb_{0.2}Sr_2CaCu_2O_{8+\delta}$ (PbBi2212). Through ITS, the superconducting gap 2Δ = 75 meV (at 10 K) is obtained for a PbBi2212 crystal. The large 2Δ value corresponds to the underdoped property of Pb-free Bi2212, which is consistent with the *ab*-plane transport measurement results performed simultaneously. The normal tunneling resistance R_N derived from the high bias region of the *I–V* characteristics is significantly small in comparison with underdoped Bi2212. Moreover, J_c of PbBi2212 is less deviated from the Ambegaokar–Baratoff value than the case of underdoped Bi2212. It is interpreted that the Pb substitution makes the tunnel barrier lower, resulting in a reduced anisotropy and a high J_c even with a lower doping.

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1. Introduction

The large anisotropy is one of the characteristic features of high-temperature superconductors (HTSC). HTSCs with a particularly high anisotropy parameter up to $\gamma \sim 1000$ are referred to as an intrinsic Josephson junction (IJJ) system because the *c*-axis transport properties are interpreted in the terms of the losephson effect [1]. This feature is commonly observed in Bi₂Sr₂CaCu₂O₈₊₈ (Bi2212). It is also known that partial substitution of Bi with Pb in Bi2212 decreases γ drastically with the critical *c*-axis current density I_c higher and the *c*-axis conductivity σ_c higher by more than one order of magnitude [2,3]. It is tacitly supposed that substitution of Bi³⁺ with Pb²⁺ in the block layer induces a hole in the CuO₂ superconducting layer and contributes to an increase in the superfluid density. However, a systematic understanding for both inter-layer (*c*-axis) and intra-layer (*ab*-plane) transport properties in PbBi2212 remains unclear despite of its high utility for IJJ devices and wire applications.

Intrinsic tunneling spectroscopy (ITS) is a powerful technique to probe quasiparticle density of states inside the crystal [4–6]. This is in contrast to the surface probing techniques like scanning tunneling spectroscopy (STS) and angular-resolved photoemission spectroscopy (ARPES), which are seriously suffered from surface deteriorations. Recently, Kinoda et al. reported STS results on PbBi2212 [7]. They showed that the superconducting gap for high J_c samples is as large as 75 meV and spatially inhomogeneous, which is similar to the underdoped Pb-free Bi2212 [8]. Although

our group has reported ITS results on PbBi2212 mesas [9], concomitant measurements of carrier density for the same crystals employed were not performed.

In this paper, ITS and Hall measurements in single crystalline PbBi2212 are presented. It is found that with Pb substitution the inter-plane (J_c and σ_c) properties drastically change while the intra-plane properties (R_H and Δ) does not change very much. This result shows that the Pb substitution greatly reduces the tunneling barrier, resulting in drastic changes in the *c*-axis properties.

2. Experiments

2.1. Crystal growth

Single crystals of PbBi2212 were grown by the self-flux method [10]. Mixed ingredient powders with a nominal molar composition of $1/2Bi_2O_3$:PbO:SrCO_3:CuO = 1.8:0.2:2:1:2 were melted in an Al₂O₃ crucible at a temperature of 1000 °C and slowly cooled to 800 °C at a rate of 1 °C/h. Electron dispersive spectroscopy (EDS) analysis of a crystal shows that the distribution of Pb within the crystal is almost homogeneous and the composition is $Bi_{1.9}Pb_{0.1}Sr_{2.1}Ca_{0.8}Cu_{2.0}O_{8+\delta}$. The *c*-axis lattice constant was found to be 3.09 nm and the bulk critical temperature T_c was obtained as 85 K by the four-probe resistance measurement.

2.2. ab-Plane transport measurement

A single crystal with flat surfaces (*ab*-plane) was taken from a congregated lump. Hall-bar structures glued on a sapphire substrate were prepared by photolithography and etching



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Fig. 1. A photograph of sample H-1 (a) and schematic illustration of the Hall-bar pattern (b).

(Fig. 1). Crystals of PbBi2212 were taken from the same batch as crystals used for ITS measurements. The Hall bar structures were fabricated from mechanically cleaved crystals with a thickness less than 500 nm. The Hall-bar pattern was formed by photolithography and either by wet etching with diluted nitric acid (Sample H-1) or by Ar-ion milling. Silver electrodes were formed by evaporation and subsequent wet etching by KI solution.

Hall measurements were performed by using the standard bridge of the Physical Properties Measurement System (PPMS) (Quantum Design Inc.) and the values for the Hall coefficient R_H were estimated by the least squares linear fitting to the Hall (transverse) voltage generated by sweeping values for magnetic field between ±1 T at a fixed temperature.

2.3. ITS measurements

Ultra-thin mesa structures (Fig. 2a) were fabricated on a crystal glued on a sapphire substrate. To reduce the contact resistance between the electrodes and the topmost IJJs, a high value of which always disturbs the precise measurement in this technique, we deposited an Ag thin film on the fresh surface cleaved inside the vacuum chamber (cleaving-in-vacuum method) [11]. The details of mesa-fabrication are described elsewhere [12,13]. The covered surface of the crystal is nearly free from deterioration due to vapor or solvents during the subsequent fabrication processes. The mesa structures were formed by electron-beam lithography and Ar-ion milling. A high resolution and a slow etching rate are necessary to make small volume mesas which enable precise measurement by reducing self-heating. Silver electrodes and SiO₂ insulation were formed by the lift-off and the self-align methods, respectively.

The *I–V* characteristics were recorded by taking traces on an analog oscilloscope under biasing triangular ac voltage with constant amplitude to the circuit in which the mesa and a 100 Ohm resistor are connected in series. ITS measurements were done by short-pulse bias method to reduce self-heating [14]. The circuit for the measurement is represented in Fig. 2b. Voltage values were acquired at ~1 µs from the pulse rise and then smoothed to give numerical differential dI/dV-V curves.

3. Results and discussion

Fig. 3a and b show temperature dependences of the *ab*-plane resistivity $\rho_{ab}(T)$ and the Hall coefficient $R_H(T)$ for a representative Hall-bar sample H-1 which has the thickness d = 60 nm. With decreasing temperature, $\rho_{ab}(T)$ decreases linearly and start to deviate from the *T*-linear behavior around 150 K. The T_c is found to be 86 K by taking the midpoint of the resistive transition. $R_H(T)$ shows a broad peak around 120 K associated with *T*-linear behavior in a higher temperature range and a rapid decrease in a lower temperature range. Comparing with data in Pb-free Bi2212 previously obtained by our group, ρ_{ab} and R_H values of these PbBi2212 sam-



Fig. 2. Schematic illustration of mesa structure (a) and diagram of the circuit for short-pulse ITS (b).



Fig. 3. *T*-dependence of the *ab*-plane resistivity ρ_{ab} (a) and the Hall coefficient R_H (b) for sample H-1.

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