

***c*-Axis Superfluid Response and Quasiparticle Damping of Underdoped Bi:2212 and Bi:2201**

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(Received 10 May 1999)

By sweeping the microwave frequency, we measured the Josephson plasma resonance for underdoped Bi:2212 and Bi:2201. The resonance enables us to determine the superfluid density and quasiparticle conductivity σ_c^{qp} in the *c* axis accurately. In the slightly underdoped crystals, superfluid density shows very little change at low temperature, which is consistent with a *d*-wave coherent tunneling model. The *c*-axis superfluid response shows an unusual doping dependence. The *T* dependence of σ_c^{qp} is very different from that in the *ab* plane. The Josephson coupling energy in single-layer Bi:2201 is a factor of 5000 smaller than predicted by the interlayer tunneling model.

PACS numbers: 74.25.Nf, 74.50.+r, 74.72.Hs

The interlayer electron transport is one of the most important subjects for understanding the mechanism of high-*T_c* superconductors (HTSC). In the normal state of HTSC, the transport properties within the superconducting planes are very different from the transport normal to the planes. A typical example is the dc resistivity of underdoped HTSC, which is metallic in the *ab* plane while it is semiconducting in the *c* axis [1]. This behavior seems to be universal in HTSC, and is an indication of the breakdown of the Fermi-liquid theory. In the superconducting state, the electrodynamics within the *ab* planes have been studied extensively, revealing key features of the *d*-wave superconducting state [2–4]. On the other hand, the *c*-axis transport of Cooper pairs and quasiparticles is not well understood, although it has been investigated both theoretically [5–11] and experimentally [2–4,12–20]. To study *c*-axis electrodynamics, the detailed knowledge of the superfluid response and quasiparticle conductivities is essential. However, obtaining information about these quantities in the *c* axis is very difficult technically due to the high anisotropy of HTSC as was pointed out in Ref. [14]. In fact, the published data about the *c*-axis penetration depth λ_c lack consistency.

Josephson plasma resonance (JPR) is a novel tool that provides important information on both the superfluid and the low-energy excitations out of the condensate [11,13,18,21,22]. Josephson plasma is a Cooper pair oscillation mode through the insulating layer. In HTSC a very sharp and stable plasma mode appears in the *c* axis because the plasma excitation energy is much smaller than the superconducting energy gap Δ [23,24]. The superiorities of JPR are as follows: First, the plasma frequency ω_{p1} ($= c/\sqrt{\epsilon_0}\lambda_c$, ϵ_0 is the dielectric constant) provides a very direct measurement of the *c*-axis superfluid density n_c via $\omega_{\text{p1}}^2 = 4\pi n_c e^2/\epsilon_0 m^*$, where m^* is the effective mass of the electron. Second, the resonance line width $\Delta\omega_{\text{p1}}$ gives a direct measurement of the *c*-axis quasipar-

ticle conductivity σ_{qp}^c . Third, JPR is applicable to the samples with very large anisotropies even when λ_c exceeds the crystal size, because the resonance occurs uniformly in the whole crystal, responding only to the electron motion in the *c* axis.

In this paper, by performing JPR experiments, we present new data about the detailed *T* dependence of n_c and σ_{qp}^c for underdoped Bi:2212 and Bi:2201. Since ω_{p1} of both members fall into the microwave window, precise determinations of n_c and σ_{qp}^c are possible compared to the optical measurements [18]. Until now, almost all JPR experiments for both Bi:2212 and Bi:2201 have been done in the cavity resonator by reducing ω_{p1} by the external magnetic field [21,22]. However, both ω_{p1} and $\Delta\omega_{\text{p1}}$ are strongly influenced by the pancake vortex alignment along the *c* axis [24]. Therefore, in the present study we developed a new technique and performed the JPR experiments in a zero field by sweeping the microwave frequency continuously. We show that the superfluid fraction $n_c(T)/n_c(0)$ is consistent with a *d*-wave coherent tunneling model. In addition, the superfluid response reveals an unusual doping dependence. The *T* dependence of σ_c^{qp} is very different from that in the *ab* plane. We also discuss the interlayer tunneling model for the mechanism of superconductivity by single-layer Bi:2201 [5,6,16,17].

Underdoped Bi:2212 and Bi:2201 single crystals are grown by the traveling floating zone method. The transition temperatures are 82.5, 77.2, and 68.0 K for Bi:2212 and 16.5 K for Bi:2201. The transition widths are less than 1.5 K. The microwave frequency was swept continuously from 20 to 150 GHz using backward-wave oscillators. The sample was placed at the center of the broad wall of the waveguide in the traveling wave TE₀₁ mode. In our configuration, we picked up only the longitudinal plasma mode which is sample size independent. We used a bolometric technique to detect very small microwave absorption by the sample. To keep the microwave power

constant at the sample position when sweeping frequency, we employed a leveling loop technique taking a good impedance matching to the waveguides. We carefully check the power level by the reference sample (thin gold plate) which is placed in the vicinity of the sample.

Figure 1(a) depicts the JPR as a function of frequency for strongly underdoped Bi:2212. Outside the resonance the microwave absorption power is almost perfectly frequency independent. The resonance line becomes broad in the vicinity of T_c because ω_{p1} goes to zero rapidly at T_c . We therefore measured the JPR by changing temperature at a constant frequency near T_c [Fig. 1(b)] [25]. These complementary measurements allow us to determine ω_{p1} and $\Delta\omega_{p1}$ accurately. The microwave absorption P_{abs} is determined by the imaginary part of the dielectric function $\varepsilon_c(\omega)$; $P_{\text{abs}} \propto \text{Im}1/\varepsilon_c(\omega)$. When $\hbar\omega_{p1} \ll \Delta$, $\varepsilon_c(\omega)$ can be expressed as $\varepsilon_c(\omega) = \varepsilon_0[1 - \omega_{p1}^2/\omega^2 - \omega_{qp}^2/\omega(\omega + i/\tau)]$, where ω_{qp} and τ are the plasma frequency and the scattering time of the quasiparticles, respectively. When $\omega\tau \ll 1$, P_{abs} can be written as

$$P_{\text{abs}}(\omega, T) \propto \frac{4\pi\sigma_{qp}(T)/\varepsilon_0}{[1 - \omega_{p1}^2(T)/\omega^2]^2 + [4\pi\sigma_{qp}(T)/\varepsilon_0\omega]^2}, \quad (1)$$

where $\sigma_{qp} = \varepsilon_0\omega_{p1}^2 e^2\tau/4\pi$ is the quasiparticle conductivity [11,26]. The resonance occurs at $\omega = \omega_{p1}$ and the line width is proportional to σ_{qp}^c .

Figure 2 shows the T dependence of ω_{p1} determined from the fitting by Eq. (1). Since ω_{p1} is much larger than $\Delta\omega_{p1}$, the peak position almost exactly coincides with ω_{p1} , and the precise determination of ω_{p1} is pos-

sible. When going from slightly to strongly underdoped Bi:2212, $\omega_{p1}/2\pi$ at $T = 0$ falls from 125 to 68 GHz, which corresponds to λ_c from 150 to 300 μm . Here we used $\varepsilon_0 = 6$ [27]. Since these λ_c are comparable to the crystal size, they are very difficult to determine by the standard methods. The c -axis critical current j_c is also obtained through the relation $\omega_{p1}^2 = 8\pi^2cdj_c/\varepsilon_0\Phi_0$, where d is the interlayer distance ($d = 1.2$ nm) and Φ_0 is the flux quantum. We find that the above ω_{p1} corresponds to j_c from 900 to 270 A/cm². Using Bi:2212 fabricated in the mesa structures, several groups have determined j_c by I - V characteristics, but it appears that the j_c value and its T dependence strongly depend on the fabrication process, shape, and areas of the junction [20,28]. On the other hand, we found that both ω_{p1} and $\Delta\omega_{p1}$ are independent of sample shape and thickness.

We first analyze $j_c(0)$ in accordance with the simplest tunneling model which assumes Fermi liquid and fully incoherent tunneling (parallel momentum of Cooper pairs not conserved). In such a model, $j_c(0)$ is given by $j_c(0) = \pi\Delta(0)/2ed\rho_c$, where ρ_c is the tunneling resistance when the superconductors are in the normal state [29]. Obviously, this relation cannot be applied to HTSC, because ρ_c in the normal state shows a semiconducting behavior. Nevertheless, several groups suggest that j_c appears to be well expressed by ρ_c just above T_c [12,16]. However, if we apply the above expression to Bi:2212 with $T_c = 82.5$ K [$j_c(0) = 900$ A/cm²] using $\Delta(0) = 25$ meV from STM measurement, ρ_c is estimated to be 370 Ω cm. This value is approximately 25 times larger than ρ_c just above T_c (16 Ω cm). If we

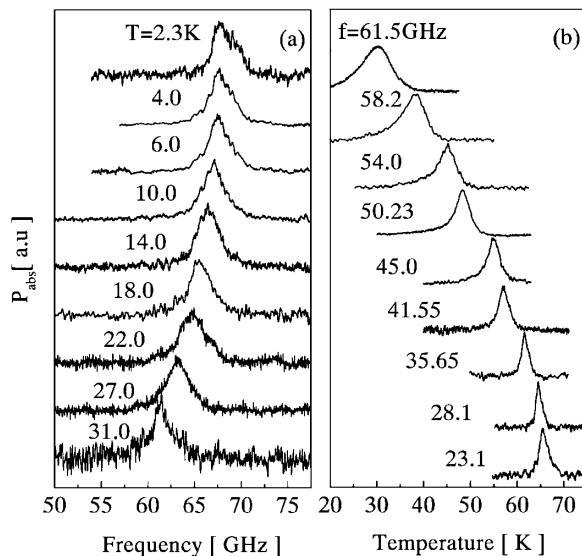


FIG. 1. (a) JPR as a function of microwave frequency for strongly underdoped Bi:2212 ($T_c = 68.0$ K). The scale of the resonance intensity was normalized by the data taken at the lowest temperatures. (b) Resonance near T_c taken by sweeping T at constant frequencies.

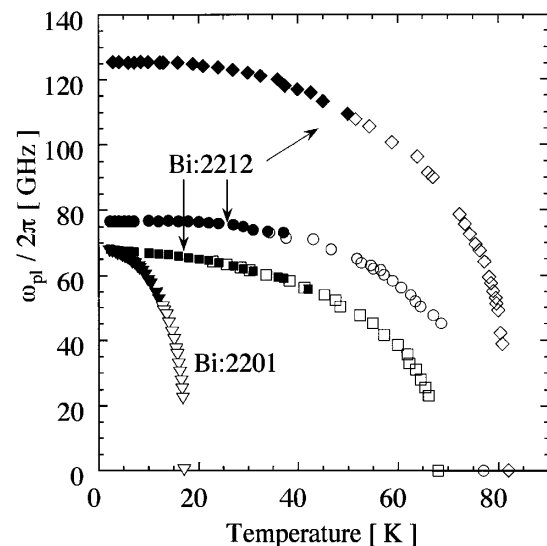


FIG. 2. T dependence of ω_{p1} for Bi:2212 and Bi:2201. The solid (open) symbols represent ω_{p1} determined by sweeping frequency (temperature). ω_{p1} determined by sweeping frequency exactly coincides with those determined by sweeping T . The temperature at which ω_{p1} goes to zero well coincides T_c determined by magnetization.

assume $\Delta = 3.5k_B T_c$ and $\varepsilon_0 = 6$ for underdoped Bi:2201, we obtain $\rho_c = 240 \Omega \text{ cm}$, which is again more than 20 times larger than ρ_c just above T_c (20 $\Omega \text{ cm}$). These results show that j_c is much smaller than expected from the normal state resistivity, and strongly indicate that the transport mechanism through the Josephson junction in HTSC is quite different from those in ordinary junctions.

We next discuss the T dependence of the superfluid fraction $n_c(T)/n_c(0)$ [$= \omega_{p1}^2(T)/\omega_{p1}^2(0)$] shown in Fig. 3. Apparently these T dependencies are much weaker than those in the ab plane which vary linearly with T . This weaker T dependence has been reported in Y:123 [2], Hg:1201, Hg:1223 [4], and La:214 [15], and seems to be universal in HTSC, although there are discrepancies in the published data. Another unusual feature appears in the doping dependence, which can be seen in the inset of Fig. 3. At $T/T_c < 0.3$, $n_c(T)/n_c(0)$ shows very little change in the slightly underdoped Bi:2212 ($T_c = 82.5 \text{ K}$ and 78 K), while a remarkable T dependence can be seen in strongly underdoped Bi:2212 ($T_c = 68 \text{ K}$) and Bi:2201. It has been suggested that the weaker T dependence in $n_c(T)/n_c(0)$ is a consequence of the in-plane anisotropy of the c -axis hopping integral t_\perp which is proportional to $(\cos k_x - \cos k_y)^2$, where k_x and k_y are in-plane momenta [10]. This t_\perp originates from the wave function overlap between the Cu $4s$ orbital and the O $2p$ orbital and is a common feature of HTSC without a chain. In the superconducting state, the node directions

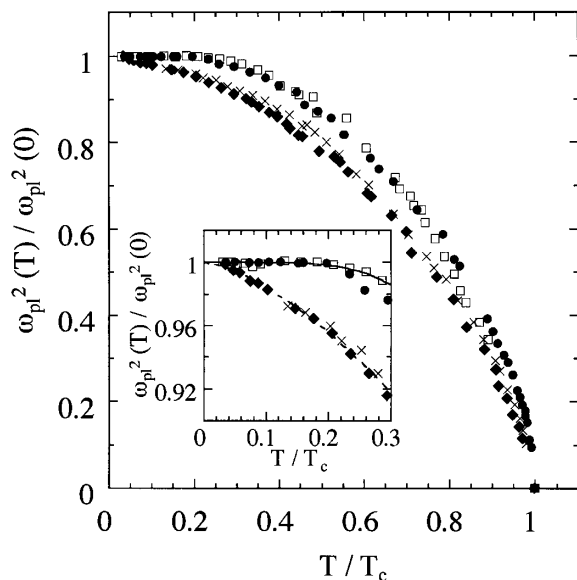


FIG. 3. Superfluid fraction vs T/T_c for Bi:2212 with $T_c = 82.5 \text{ K}$ (solid circles), 77.2 K (open squares), and $T_c = 68.0 \text{ K}$ (solid diamonds), and for Bi:2201 with $T_c = 16.5 \text{ K}$ (crosses). The inset shows the same data at low temperatures. The solid line represents $\omega_{p1}^2(T)/\omega_{p1}^2(0) = 1 - A(T/T_c)^\alpha$, with $A = 6$ and $\alpha = 5$. The broken line represents $\omega_{p1}^2(T)/\omega_{p1}^2(0) = 1 - B(T/T_c)^\beta$ with $B = 0.5$ and $\beta = 1.5$.

of the $d_{x^2-y^2}$ symmetry and zeros of t_\perp coincide. This weakens the d -wave node contribution and leads to a weaker T dependence of n_c .

In the d -wave superconductor, j_c due to the simple diffusive (fully incoherent) tunneling process vanishes, resulting from the vanishing average of the d -wave order parameter in momentum space [8]. The finite j_c arises from two conduction channels: the incoherent process with anisotropic impurity scattering [impurity-assisted hopping (IAH)] and the coherent process (parallel momentum conserved) [7]. When coherent, it is predicted that n_c varies as T^5 at low temperatures [10]. On the other hand, the IAH model predicts that n_c varies quadratically with T . This is because the anisotropic impurity scattering enhances the contribution of the node directions, which in turn enhances the T dependence of n_c . The T^5 dependence has been observed in low anisotropic Hg:1201 [4]. For slightly underdoped Bi:2212, $n_c(T)/n_c(0)$ is well expressed as $1 - A(T/T_c)^\alpha$ with $\alpha = \sim 4-6$ and $A \sim 6$. These α and A are close to the values reported in Hg:1201. Thus almost T -independent $n_c(T)/n_c(0)$ in slightly underdoped Bi:2212 is in disagreement with the IAH model, suggesting that *even in extremely anisotropic Bi:2212 the coherent transport plays an important role in determining the superfluid response*. This result is consistent with the recent I - V measurements which showed that the tunneling process is coherent in Bi:2212 [20]. These results imply that the impurities act as strong scattering centers in the interlayer tunneling process, because in the strong limit the scattering becomes isotropic and the contribution of IAH becomes negligibly small. On the other hand, $n_c(T)/n_c(0)$ for strongly underdoped Bi:2212 and Bi:2201 at low temperatures are well fitted by $1 - B(T/T_c)^\beta$ with $B \sim 1$ and $\beta = \sim 1.5-2$. This T dependence is close to the prediction of the IAH model [4,10]. Therefore the change of the T dependence of n_c with doping may be due to a crossover from coherent to IAH. However, it seems to be unlikely that such a crossover occurs in the extremely anisotropic Bi:2212. Thus the origin of the T^2 dependence of $n_c(T)$ may be due to other mechanisms which enhance the node contribution. To clarify this point, the detailed knowledge of the electronic structure in strongly underdoped regime is necessary.

Figure 4 shows σ_{qp}^c obtained from $\Delta\omega_{p1}$ by the fitting of Eq. (1) for underdoped Bi:2212 and Bi:2201. Small but finite inhomogeneous distribution of T_c in the sample may also broaden the resonance line. However, we have measured several crystals with the broader transition widths and observed the same $\Delta\omega_{p1}$. We therefore believe that σ_{qp}^c is a main factor for $\Delta\omega_{p1}$. Below T_c , σ_{qp}^c decreases gradually with decreasing T . The rapid decrease of σ_{qp}^c just below T_c from the dc value may be due to the two fluid analysis [Eq. (1)] which is not a good approximation near T_c . Similar T dependence of σ_{qp}^c was observed in all samples. At low temperatures, σ_{qp}^c of both members remains finite. This fact can also

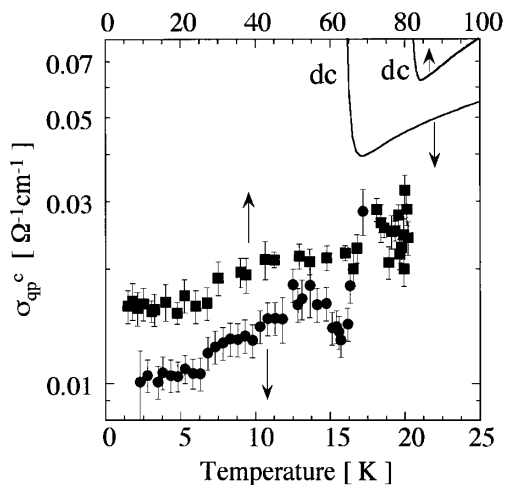


FIG. 4. Quasiparticle conductivity for slightly underdoped Bi:2212 with $T_c = 82.5$ K (solid squares) and Bi:2201 with $T_c = 16.5$ K (solid circles). The c -axis dc conductivities above T_c are also shown.

be confirmed by the integrated intensity of the resonance that also remains finite at low temperatures (see Fig. 1). This is because in d -wave superconductors the impurity scattering gives rise to a finite quasiparticle density of states at the Fermi level. The monotonic decrease of σ_{qp}^c below T_c is in contrast to the quasiparticle conductivity in the ab plane which shows a broad peak below T_c due to the suppression of the quasiparticle scattering [2,3]. This suggests that the quasiparticle transport in the c axis is not influenced by that in the ab plane, similar to Y:123 with low anisotropy [14].

Finally, we would like to include some comments about the interlayer tunneling (ILT) model which predicts that the superconducting condensation energy E_c is approximately equal to E_J ; $E_c = \eta E_J$ with $\eta \approx 1$ [5,6]. The single-layer superconductors pose a rigorous test for the ILT model. It has been suggested that the prediction holds for La:214, but it is strongly violated in Hg:1201 ($\eta \sim 50$) [16] and Tl:2201 ($\eta \sim 400$) [17,18]. However, it has recently been pointed out that the conclusion regarding the violation of the ITL model is premature because the estimation of E_c for Hg:2201 and Tl:2201 has numerical ambiguity [6]. In single-layer Bi:2201 we obtain $E_J [= \epsilon_0(\hbar\omega_{pl}/2ed)^2/4\pi] \approx 7.3$ erg/cm³ from $\omega_{pl}/2\pi = 68$ GHz. Unfortunately, there is no specific heat data of the Bi:2201. If we assume that E_c of Bi:2201 is close to E_c of La:214 with the same T_c and that E_c is proportional to T_c^2 , E_c is estimated to be $\sim 4.3 \times 10^4$ erg/cm³ [5]. This indicates $\eta \geq 5000$ for our Bi:2201. In other words the ILT model predicts that E_c of La:214 is more than 5000 times larger than E_c of Bi:2201 with the same T_c . This is quite unlikely, although the assumption used for this estimation may be rough. Thus our results provide additional strong evidence that

the Josephson coupling is not relevant to the pairing interaction [16–18].

In summary, we have measured JPR in a zero field in underdoped Bi:2212 and Bi:2201. The T dependence of the superfluid fraction is consistent with the d -wave coherent tunneling model with strong scattering. The T dependence of quasiparticle conductivity in the c axis is quite different from that in the ab plane. In single-layer Bi:2201, the Josephson coupling energy is a factor of 5000 smaller than predicted by the interlayer tunneling model.

We gratefully acknowledge discussion with P. W. Anderson, N. E. Hussey, M. Imada, T. Koyama, N. Nagaosa, M. Ogata, N. P. Ong, S. Tajima, and Y. Tanaka and the critical reading of the manuscript by L. N. Bulaevskii.

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