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1. Introduction

Comparing with normal states, the electromagnetic responses of superconductors below the critical temperatures (T_c) should change considerably since the emergence of Cooper pairs. According to the sum rule, one learns that the loss of the spectra weight in high energy regions transfers to a dc δ function (corresponding to the dc zero resistivity), making a conservation of the whole area of $\sigma_1(\omega)$, the real part of the complex conductivity [1–6]. Strictly speaking, an ideal zero frequency could not be realized in experiments, so one may expect that $\sigma_1(\omega)$ would diverges in the dc limit from a finite value at finite frequencies in superconducting states. Naturally, investigating the detailed behavior of the divergence or the evolution of $\sigma_1(\omega)$ in low frequency regions near T_c will be helpful and necessary to understand the process of superconducting phase transition and the sum rule.

Up to now, much less research work has been done in lower frequency regions $(10^{0}-10^{8} \text{ Hz})$ for correlated electron systems or high- T_{c} cuprates, though it probes the bulk properties and provides experimental access to the dynamical properties even at $T < T_{c}$ in such a wide frequency range. The reason may be that in superconducting states, the supercurrent would make it difficult to detect a rather weak but finite electrical transport signal. Additionally, the parasitic effect could not be neglected above 10^{6} Hz. However, in

ABSTRACT

We report the in-plane temperature dependent ac impedance spectrum of Tl₂Ba₂CaCu₂O_{8+δ} (Tl-2212) single crystal film with thickness 3000 Å and T_c = 96 K. In process of the superconducting condensation, the impedance spectrum $z(\omega, T_0)$ changed considerably and was very similar with the evolution of the spectrum under various dc bias voltages in the shallow superconducting state. The spectrum weight transfers from high frequency regions to lower ones and eventually diverges towards the dc limit. From the analysis, it seems that the superconducting 'islands' form first and then grow larger, till the formation of a 'continent'. The impedance spectrum $z(\omega, B_l)$ at various dc bias B_l have also been studied.

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a shallow superconducting state or when the superfluid density ρ_s has been suppressed partly, the temperature dependent ac responses in low frequency range could be identified well [7–9]. Such results will be undoubtedly necessary to understand the phase transitions and electrodynamics of correlated electron matters or high- T_c cuprates. In experiments, the electrical properties in low frequency region up to 10^8 Hz could be probed by circuit techniques, while for higher frequency regions, the microwave or optics approaches are often used.

In present work, the ac impedance spectrum $z(\omega,T_0)$ of the specimen Tl₂Ba₂CaCu₂O_{8+ δ} film (Tl-2212) has been investigated in the superconducting condensation process and further more, the impedance spectrum $z(\omega,B_l)$ at various dc bias B_l have also been studied, the dc bias current is used to deliberately tune ρ_s in shallow superconducting states. Here T_0 is lightly less than T_c . In the process of the superconducting phase transition, the spectrum evolves continuously and eventually diverges towards the dc limit. In shallow superconducting states, as ρ_s has been tuned, the corresponding impedance spectrum changes remarkably. The evolution of the responses in low frequency regions could be attributed to the formation of superconducting 'islands'. Below T_c , the impedance spectrum has been suppressed as ρ_s increases.

2. Experiments

The samples in our experiments are Tl-2212 epitaxial single crystal films with typical thickness 3000 Å on LaAlO₃(001) substrates, details of the films fabrication are described elsewhere



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Fig. 1. The dc and ac temperature-dependent resistivity of TI-2212 single crystal film with $T_c = 96$ K. Note that the two curves are nearly identical with each other. The triangles represent the temperatures at which the impedance spectra are probed. Upper inset: the measurement configuration of dc conductivity. Lower inset: The X-ray diffraction pattern up to 60° for a TI-2212 film (LaAlO₃(001) substrate). Note that the (00*l*) peaks are clearly identified, indicating a good quality of the sample.

[10]. From the XRD results carried out on Rigaku D/Max-2500X, the samples have good orientations, see the inset of Fig. 1. All the films have shiny planes and about 3 mm \times 2 mm surface sizes. Epoxy H20E silver paint electrodes are applied on the specimens and annealed at 623 K for 30 min to make high quality of contact. The dc temperature dependent resistivity has been measured using Keithley-2812 source meter with a constant current *I* = 10 μ A and Keithley-2400 nanovolt meter, see the upper inset of Fig. 1. The



Fig. 2. (a) The fittings to the impedance spectrum of Tl-2212 film at various temperatures in the normal states. (b) The temperature dependent resistance (R) and parasitic inductance (L_p). Inset: the effective circuit model used in fittings.



Fig. 3. (Upper left) the fittings to the impedance spectra of TI-2212 film in the process of the superconducting condensation at various temperatures. (Upper right) the calculated impedance spectra in the process of the superconducting condensation with the superfluid electrons having a homogeneity distribution. Insets: the effective circuit model used in fittings. Note that R_N presents the region of the normal components, whereas R_S and L_S presents the superconducting islands. R_R presents for the resolution limit. (Lower) the schematic for the evolution of the superconducting phase transition from the discrete 'islands' to a 'continent' superconducting state eventually (a–c).

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