

## Partial Destruction of the Meissner State in Multiconnected Josephson Network

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In the course of superconducting transition of multi-connected superconductor, we first observed peculiar instabilities of ac fundamental susceptibility  $\chi_1' - i\chi_1''$  and third-harmonic susceptibility  $|\chi_3| = (\chi_3'^2 + \chi_3''^2)^{1/2}$ . It is suggested that the quantized flux shooting is responsible for the observed instability.

KEY WORDS: Superconductivity/ ac susceptibility/ Third-harmonic susceptibility/ Josephson network/

### INTRODUCTION

The possibility of cooperative behavior in the superconducting system consisting of a number of weak links has been investigated with considerable interest. Recently, we studied the response of superconducting Tc with multiconnected structure against small ac magnetic field  $h(t) = h_0 \sin \omega t$  in terms of ac fundamental susceptibility  $\chi_1' - i\chi_1''$  and its higher harmonics  $|\chi_n| = \sqrt{\chi_n'^2 + \chi_n''^2}$ .<sup>1,2)</sup> There, we proposed a weakly-connected loop model, *i.e.*, the specimen is regarded as a random network of Josephson weak links and a number of loops excited in the network against external magnetic field are assumed to behave like a single loop as a whole owing to the coherent nature of the system. This model well explains the observations; the  $h_0$ -sensitive and  $T$ -dependent properties of fundamental susceptibility and the variation of higher-harmonic susceptibilities against temperature.

Previous results also contained an interesting fact, *i.e.*, the peculiar instability in susceptibility curves of superconducting transition ( $h_0 > 2$  Oe) was observed for the first time. The purpose of the present work is to give a brief discussion on the origin of this partial destruction of the Meissner state.

### EXPERIMENTAL

The details of sample preparation and measuring system were previously reported.<sup>1)</sup> A sample was prepared by reducing  $\text{NH}_4\text{TcO}_4$  embedded in a porous alumina. First, an aqueous solution of  $\text{NH}_4\text{TcO}_4$  was soaked in the porous alumina and it was evaporated to dryness at  $80^\circ\text{C}$  in flowing  $\text{N}_2$  gas. The reduction procedure was performed in two steps ( $300^\circ\text{C}$  for 2 h and  $600^\circ\text{C}$  for 3 h) in the  $\text{H}_2$  atmosphere.

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After the reduction, the specimen was annealed in the atmosphere of Ar containing a few %  $H_2$  at  $1000\sim 1200^\circ C$  for 6 h.

In the specimen, a lot of microbridge-like structures of Tc metal were found among alumina particles. This means that the specimen consists of multiconnected Josephson network.

The measuring system of  $\chi_1'$  and  $\chi_1''$  was the Hartshorn-type ac bridge. For the measurements of  $|\chi_3|$  and  $\chi_1''$ , the system was modified, where a lock-in analyzer coupled with Lissajous figure was used to pick up selectively the third harmonic in ac response. The temperature of specimen was measured by a Ge thermometer.

## RESULTS AND DISCUSSION

### 1. Fundamental and Third-Harmonic Susceptibilities

By using the Hartshorn bridge, ac susceptibility of the specimen was measured in respect of temperature  $T$  as well as of the amplitude of external magnetic field  $h_0$ . In Fig. 1 are shown the typical results for external magnetic field of 40 Hz. As seen in the figure, the experimental curves are characterized in two respects: (1) The transition width  $\Delta T$  of  $\chi_1'$  ( $10\sim 90\%$ ) is sensitively broadened as  $h_0$  increases, but the onset temperature is not altered. And the curve  $\chi_1''$  forms a peak. These properties have been well explained by means of the weakly-connected loop model.<sup>1)</sup> (2) Observed curve at  $h_0=2.70$  Oe shows the peculiar instability of the superconducting Meissner state near the completion of the transition and its recovery at lower temperature. Namely, at  $6.92\sim 6.94$  K, both  $\chi_1'$  and  $\chi_1''$  show rapid switching between the

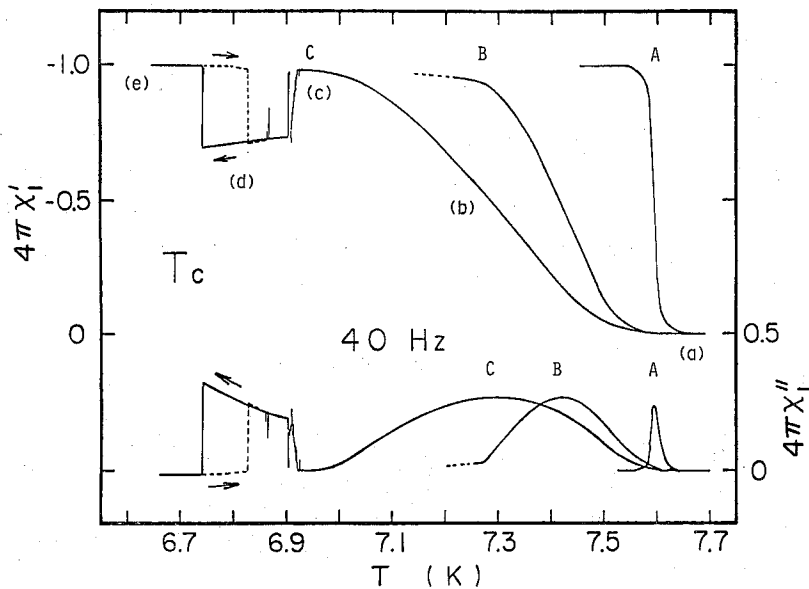


Fig. 1.  $\chi_1'$  and  $\chi_1''$  of a multiconnected Tc specimen *vs.* temperature for various amplitudes of ac field. (A) 0.00296 Oe, (B) 1.22 Oe, (C) 2.70 Oe.

superconducting state and the intermediate (superconducting-normal) state. At 6.90~6.75 K, they stay in the intermediate state and change gradually. Below 6.74 K, abrupt recovery of the superconducting state takes place, and  $\chi_1'$  and  $\chi_1''$  are constant down to the lowest temperature of our measurement, 1.23 K. Both  $\chi_1'$  and  $\chi_1''$  have a hysteresis with respect to temperature around 6.8 K.

Simultaneous measurements of  $|\chi_3|$  and  $\chi_1''$  were also performed with respect to  $T$  as well as to  $h_0$ . In Fig. 2 are shown the typical results for external magnetic field of 41.5 Hz. Results are summarized as followings: (1) As  $h_0$  increases, the temperature region of  $|\chi_3|$  transition is broadened and this is in close correlation with  $\chi_1''$ . The proportionality of  $|\chi_3|$  to  $\chi_1''$  is obvious. Our model has well reproduced this feature of observations.<sup>1)</sup> (2) The instability similar to that of fundamental susceptibility, as noted above, is found in the curve C at temperature near 6.8 K.

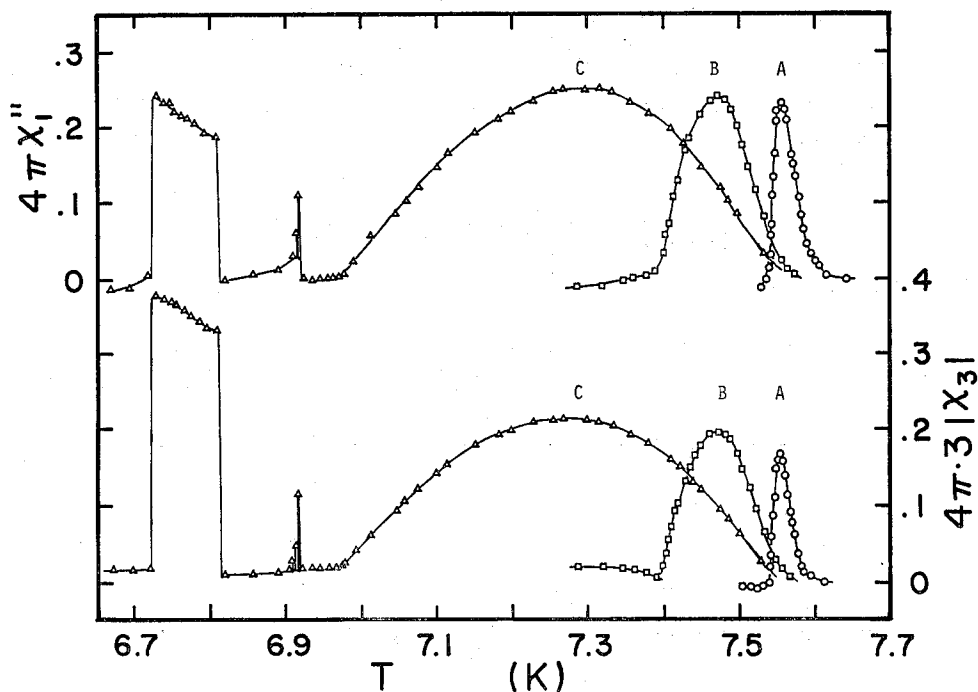


Fig. 2.  $\chi_1''$  and  $|\chi_3|$  of a multiconnected Tc specimen vs. temperature. The amplitudes of ac field are (A) 0.0488 Oe, (B) 0.532 Oe, (C) 2.71 Oe. Fundamental frequency is 41.5 Hz.

## 2. Off-Balance Signal

Since the measurements of  $\chi_1'$ ,  $\chi_1''$ , and  $|\chi_3|$  were carried out through the lock-in analyzer, the harmonics in response were selectively picked up. But the complicated behavior of susceptibility indicates its non-linear response against  $h_0$ . Therefore, off-balance signals of the bridge are expected to contain the detailed informations, which probably offer the key to the problem.

Observed wave forms of off-balance signals  $e(t)$  by an oscilloscope in ac magnetic field ( $h_0=2.78$  Oe) are schematically reproduced in Fig. 3, where (a)~(e) correspond

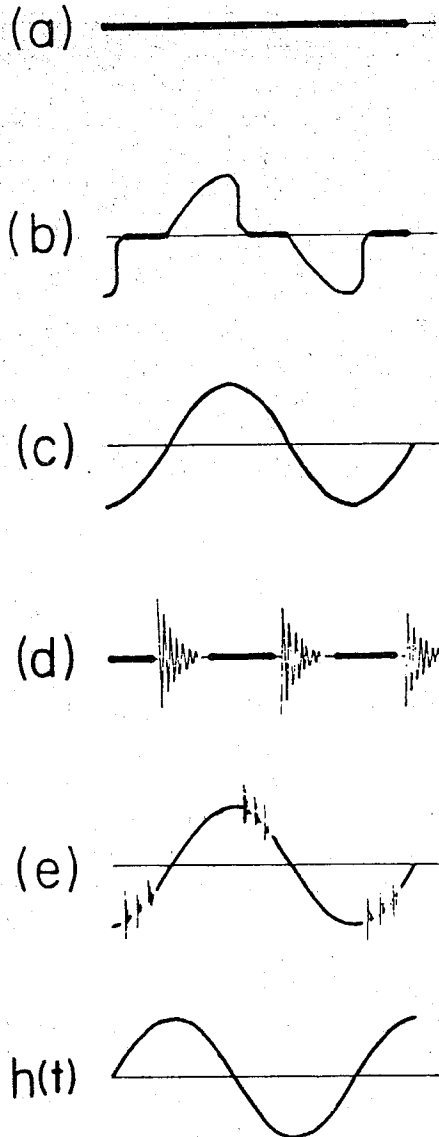


Fig. 3. Schematic wave forms of observed off-balance signal  $e(t)$ .  $h_0$  is 2.78 Oe. Temperatures of the specimen are (a) 7.641 K, (b) 7.259 K, (c) 6.945 K, (d) 6.837 K, (e) 5.093 K. (a)~(e) correspond to points indicated on curve C of Fig. 1. Vertical scale for (d) is 25 times larger than others. Applied ac field  $h(t)$  is also presented.

to points indicated in Fig. 1. Among them, (a)~(c) give one of the convincing evidences for the plausibility of the weakly-connected loop model.<sup>1)</sup> The wave form (d) has a peculiar structure, *i.e.*, periodic oscillation with rather rapid damping (see Fig. 4). This oscillation is induced every half period of  $h(t)$  and the sign is opposite to its neighbor. This remains in the wave form (e), but its amplitude is appreciably lessened.

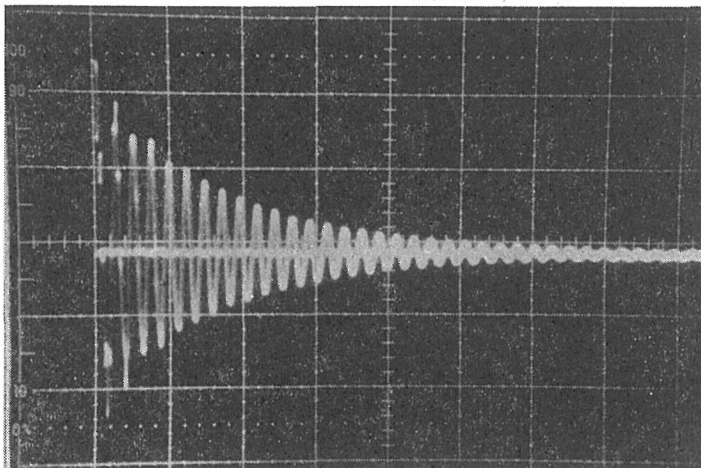


Fig. 4. Observed wave form for point (d) in Fig. 1 ( $T=6.869$  K,  $h_0=2.78$  Oe). Vertical scale is 50 mV/div and horizontal scale is 0.5 msec/div.

In the next section, we give a brief discussion of the origin of instability in connection with the results appeared in Figs. 1~4.

### 3. Instability Caused by Flux Quanta Shooting

To explain the  $T$ -dependent and  $h_0$ -sensitive features of the fundamental susceptibility and of the third-harmonic susceptibility, we proposed the phenomenological model,<sup>1)</sup> where we considered that the specimen can be regarded as a multiconnected Josephson network. By imposing small ac magnetic field upon the specimen, a number of shielding loops may be excited in the network, where flux quanta can enter or leave the loops through weak links. For a multiconnected superconducting geometry, the possibility of long-range ordering over the system against small magnetic field was pointed out by the group of Rosenblatt.<sup>3)</sup> We considered that many loops appeared in the network behave like a single loop as a whole due to the coherent nature of the specimen.

However, we paid no regard to the transient current which may be induced in the loop when a flux quantum passes through a junction involved in the loop. In other words, we assumed that the induced current is well damped due to the loop impedance and results in negligible contribution to the observed susceptibilities ( $h_0 < 2$  Oe).

It is well known that the characteristics of weak-link junctions are closely related to its ac impedance.<sup>4)</sup> In the present case, since the specimen contains a lot of microbridge-type junctions, the electromagnetic behavior is specified by introducing a series inductance seen from the active junction in an equivalent-circuit analysis.

To understand the unstable response described above, we extend our discussion to the transient current  $j(t)$  induced in the loop by traversing flux quanta. The induced voltage corresponding to  $j(t)$  by single flux quantum is expected to be impulsive. However, sequential traverses of many flux quanta over the junctions may result in

generation of a step-function of height  $V_0$ .

Assuming a superconducting loop in the dissipative state is equivalent to a series circuit of  $L$ ,  $C$ , and  $R$ ,  $j(t)$  can be classified into three cases depending on the loop impedance, where  $R$  denotes the junction resistance and  $C$  is the stray capacity. Supposing the initial charge stored in  $C$  is zero, one gets

$$\left. \begin{aligned} j(t) &= (V_0/\Omega L)e^{-\beta t} + \sin \Omega t & \text{for } R < 2(L/C)^{1/2} \\ &= (V_0/L)te^{-\beta t} & \text{for } R = 2(L/C)^{1/2} \\ &= (V_0/\gamma L)e^{-\beta t} \sinh \gamma t & \text{for } R > 2(L/C)^{1/2} \end{aligned} \right\} \quad (1)$$

where

$$\left. \begin{aligned} \beta &= R/2L \\ \Omega &= [(1/LC) - (R/2L)^2]^{1/2} \\ \gamma &= [(R/2L)^2 - (1/LC)]^{1/2} \end{aligned} \right\} \quad (2)$$

The first solution in Eq. (1) can be reasonably connected to the construction of wave form found at point (d) in Fig. 3, and the second and third solutions correspond to well damped induced current. In the following discussion, we assume  $R$  and  $C$  are constant regardless of loop dimension, *i.e.*, the applicability of three solutions given in Eq. (1) refers only to  $L$ .

At the early stage of superconducting transition, there appear small loops through which the shielding current flows. The loop inductance in this region is evidently small and the transient current  $j(t)$  is considered to be expressed by the second or third solution. By this  $j(t)$  emerged in the small loops, the abrupt change in magnetization  $m(t)$  caused by  $\phi_0$  penetration may be obscured, but this is not critical for the discussion presented in the weakly-connected loop model. When temperature is lowered, the available shielding-current density  $J_0(T)$  of each loop increases, and thus the magnetically shielded area in the specimen becomes larger, resulting in increased  $L$ .

If the whole specimen becomes in the nearly Meissner state, but  $J_0(T)$  is not sufficiently large, magnetic flux condensed near the sample edge is shot into the specimen due to a large magnetic pressure, where the quantized flux shooting takes place every half cycle of applied field, but the sign of quanta is alternative. Supposing that a number of flux quanta are subsequently shot from the sample edge, and their crossing path length is not negligible compared with the size of specimen, and appreciable amount of electromagnetic force  $V_0$  is induced in the loop. Besides, in the situation where the superconducting transition is nearly completed,  $L$  is sufficiently large. Thus we can reasonably deduce the first solution in Eq. (1). Actual observation of  $e(t)$  is proportional to  $dj(t)/dt$ . Assuming  $\beta \ll \Omega$ , one gets  $e(t) \propto \exp(-\beta t) \cdot \cos \Omega t$ , which seems to support our observation shown in Fig. 3, where the large amplitude of the oscillating signal is due to the largeness of  $V_0$ .

It should be noted here that for  $h_0 < 2$  Oe we have not observed the peculiar response. This is probably due to the small magnetic pressure at the edge of the specimen and no shooting of magnetic flux takes place.

When temperature is further lowered, the available shielding current density increases and thus long-distance flux shooting into the specimen hardly takes place.

This corresponds to the recovery of the stable diamagnetic state (see Fig. 1). However, there may be some residual short-distance shootings of a small number of flux quanta, which probably corresponds to the oscillation superimposed on sine wave [see Fig. 3(e)].

The fundamental and third-harmonic Fourier components of the peculiar damping oscillation result in the behavior of  $\chi_1'$ ,  $\chi_1''$ , and  $|\chi_3|$  shown in Figs. 1 and 2.

The discussion developed here as to the partial destruction in the multiconnected Josephson network suggests that the motion of flux quanta plays an important role in the properties of the multiconnected superconducting system and the higher-harmonic technique is a potential tool for the examination of flux motion in the system.

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