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Superconducting Transition of Bulk Granular Systems (Ta)

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Electrical resistance of the bulk granular systems consisting of Ta particles was measured in the temperature range of 1.5–300 K. It has been revealed that there is a systematic correlation between the temperature-dependent behaviors in the normal state and in the paracoherent state which appears during superconducting transition. This reflects the strength of inter-grain coupling. We also measured the complex susceptibility, which shows strong dependence on the field amplitude below a certain temperature lower than the superconducting onset. A brief explanation is given in terms of the previously proposed model.

KEY WORDS: Superconductivity/ Granular System/ Electrical Resistance/ Complex Susceptibility/

I. INTRODUCTION

Recently Josephson-coupled superconducting arrays have been found to reveal a variety of phenomena unseen in usual bulk superconductors.[1] The simplest system of this kind is granular superconductors where superconducting grains are embeded in a nonsuperconducting matrix. Perceiving this system equivalent to the three dimensional X-Y model, Rosenblatt *et al.*[2] studied the thermodynamical critical phenomena concerned. According to them, superconductivity in the granular system undergoes two-step transition, where the higher temperature transition comes from individual grains and the lower reflects a long range phase coherence between grains. In the temperature between two transitions the inter-grain Josephson-phases are thermally disordered. This is called the paracoherent state. Tsuei[3] reported Josephson-like response against microwave and flux-trappnig effect in the similar systems. Inter-grain coupling is considered to be responsible for these phenomena.

Meanwhile, so-called low dimensional superconductors have been recently got into the spotlight. For example organic superconductors,[4] $(SN)_x$,[5] transition metal calcogenides MX_3,[6] and Nb_3X_4,[7] are well known as quasi-one-dimensional synthetic materials, and graphite intercalation compounds,[8] artificially layered superconductors,[9] (intercalated) $2H\text{-}TaS_2$,[10] as quasi-two-dimensional. These fibrous or layered superconductors are known to exhibit unusual magnetic or transport properties due to strong anisotropy. Considering that the materials mentioned above are weakly coupled assemblies of one dimensional fibers or two dimensional layers, it is important to clarify the effect of inter-fiber or inter-layer coupling.

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From this point of view, we previously reported the magnetic response of quasi-one-dimensional Nb_3Se_4 in terms of the complex susceptibility, where the nature of the coupling is pointed out to be responsible for unusual dependence of the susceptibility on the field amplitude.[11] On these lines, the present work makes an attempt to investigate the electromagnetic response of weakly coupled superconducting inclusions without knotty problems involved in low dimensionality, such as charging effect[12] and fluctuation in each superconducting inclusion. As a model substance, we prepared the system composed of many superconducting particles in contact with each other, after Ref. 13. The distinctive feature of this system is controllability of inter-grain coupling (see below). In this paper we report electrical resistances of the bulk granular systems in the temperature range of 1.5–300 K to investigate the relation of the superconducting transition to the normal state resistance. The complex susceptibility was also measured to see the magnetic response. We give a qualitative discussion about the susceptibility in terms of the previously proposed model.

II. EXPERIMENTAL

Bulk granular systems are composed of superconducting tantalum particles (nominally 99.9% purity) and insulating epoxy resin (Stycast 1266). The dimension of particles is less than 44 μm in diameter, but the shape is irregular. Samples were prepared in the following manner: First, particles were mixed with a constituent liquid of the resin and stirred up for an hour or two to achieve good macroscopic homogeneity. Second, the other constituent was poured and the muddy mixture, after quickly stirred, was molded into the form of cylinder, which is 10 mm long and 2 mm in diameter. During hardening, pressure was applied with the screw bolt, where the control allowed us to get a series of samples of various resistances. After compressed at least for four days, the samples were taken out from the mold. In a measurement of electrical resistance, considerable attention should be given to the electrode. For in such an inhomogeneous medium, it is difficult to have a good contact of a lead to give a uniform current density. To make any improvement in this respect, silver was vapordeposited on both faces of the samples, and solder was put as a buffer material between the face and a copper lead. This material was examined not to serve as an additional resistance or as a thermoelectric power. The samples were mounted in the copper block with Apiezon N grease, then installed into the adiabatic room (see Fig. 1). Four-terminal electrical resistances were measured in the temperature range between 1.5 and 300 K. The temperature was controlled by means of a heater and heat exchange helium gas. For measurements above 77 K, we filled the dewar with liquid nitrogen, while below 77 K liquid helium was used. To obtain below 4.2 K, the helium bath was evacuated. A carbon glass resistor (Lake Shore, model C.G.R.-1-1000) was used as a thermometer. Prior to the experiment, its calibrated data was fitted with the following form[11]

$$\log T = \sum_{i=1}^{10} A_i \left(\ln \frac{R}{100}\right)^{i-1} \tag{1}$$

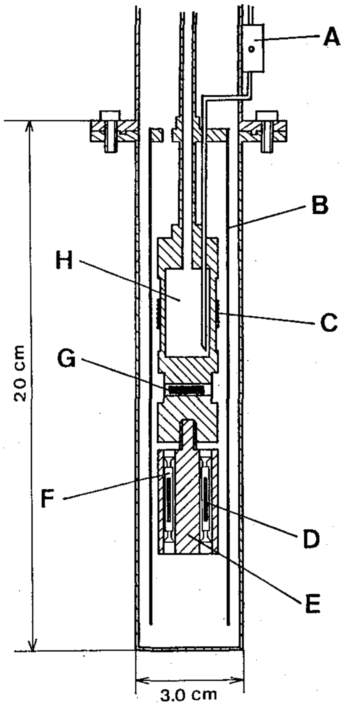

Fig. 1. Adiabatic room for measurements of electrical resistance. A: Needle valve (closed), B: Cu radiation shields, C: Heater, D: Sample, E: Cu block, F: Sample holder (lucite), G: Carbon glass thermometer, H: Helium pot (not used in the present work).

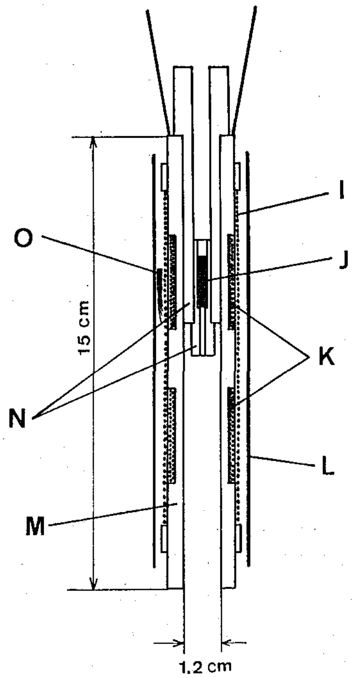

Fig. 2. Stationing of the sample and the coil for measurements of complex susceptibility. I: Primary coil, J: Sample, K: Pick-up coil, L: Coil-foil, M: Lucite bobbin, N: Sample holder (lucite), O: Ge thermometer.

where T is the temperature, R is the resistance of the thermometer, and A_i is a coefficient. Accuracy was better than 0.1% in the temperature below 100 K. The microcomputer (Hewlett Packard, model HP-85) connected with the digital voltmeter (Takeda, model TR-6877) with GP-IB displays the temperature corresponding to the resistance measured.

By the use of the Hartshorn-type mutual inductance bridge (off-balance method),[14] the complex susceptibility ($\chi = \chi' - i\chi''$) was measured with one of the samples. In Fig. 2 is shown the stationing of the primary coil, the pick-up coil, the specimen, and the thermometer. These are soaked in a liquid helium bath. For the susceptibility measurement we used a germanuim thermometer.

III. RESULTS AND DISCUSSION

1. Electrical resistance

In Fig. 3 are shown the typical results of temperature dependence of the normal state resistance. All samples show monotonic change and saturate to the value at 4.5 K. Different samples have different temperature coefficient, negative or positive. First, we must clarify what causes the change in resistance against temperature.

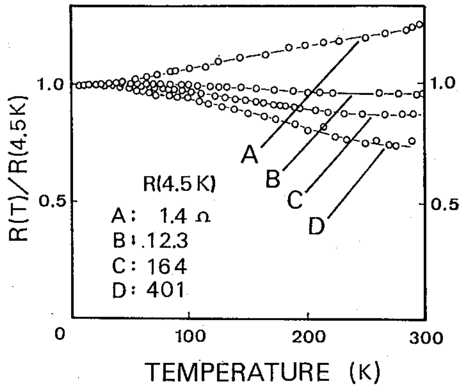

Fig. 3. Resistance vs. temperature in the normal state. Data are normalized to the value at 4.5 K. A–D indicate 4 different samples.

The measured resistance involves both the intra- and inter-grain resistances. But, considering the resistivity of tantalum is of the order of 10^{-6} Ωcm and that of the samples are greater than 10^{-2} Ωcm, we find the inter-grain resistance is evidently predominant. The temperature dependence of this resistance may be attributed to two effects; (1) thermal expansion of tantalum particles and resin, and (2) intrinsic resistance of the junction between grains. As for (1), the rate of thermal expansion for resin is ten times greater than that of tantalum. This means that the resin tends to make the praticles closer as the temperature decreases, leading to the positive coefficient. Therefore variation in the temperature coefficient comes from the junctions constituting the samples. The junctions are considered to be composed of point contacts and tunnel barriers. The former generally behaves like metallic (positive coefficient), while the latter shows negative coefficient due to thermally activated conductivity. So, in tightly compacted samples, point contacts dominate tunnel barriers.

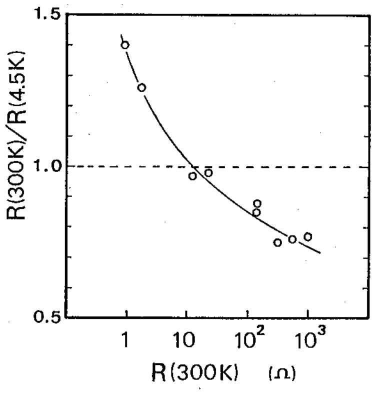

Fig. 4. Resistance ratio $R(300\ \mathrm{K})/R(4.5\ \mathrm{K})$ vs. R (300 K). Solid line guides the eyes. Dotted line represents zero temperature coefficent.

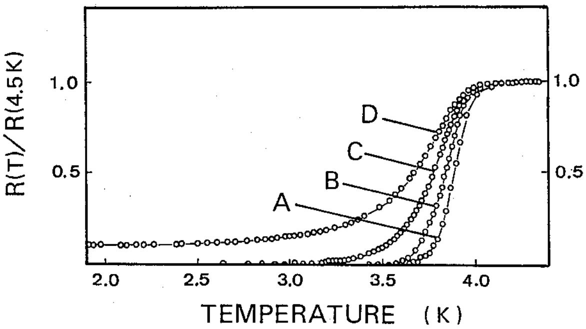

Fig. 5. Resistance vs. temperature in the superconducting transition region. Data are normalized to the value at 4.5 K. A–D correspond to samples in Fig. 3.

Besides, in Fig. 4, we see apparent correlation between the temperature dependence and the magnitude of resistance, where the resistance ratio $R(300\ \mathrm{K})/R(4.5\ \mathrm{K})$ is plotted as a function of $R(300\ \mathrm{K})$. As is seen, a critical value of $R(300\ \mathrm{K})$ is about 10 Ω, where the temperature coefficient crosses over from positive to negative.

Next, we give the superconducting transition in Fig. 5, where A-D correspond to those in Fig. 3. The onset temperature is the same, but the transition width gives rise to a remarkable variety. Compared to Fig. 3, we acknowledge the correlation between the temperature-dependent behaviors in the normal state and in the superconducting transition: As the temperature coefficient of resistance in the normal state goes to negative, the superconducting transition width is broadened. To see this correlation, we give Fig. 6, where the transition width ΔT is defined as an interval between the temperatures which give 20% and 80% of $R(4.5\ \mathrm{K})$. In case $R(300\ \mathrm{K})/R(4.5\ \mathrm{K})>1$, ΔT is nearly constant (0.15 K), while for $R(300\ \mathrm{K})/R(4.5\ \mathrm{K})<1$, ΔT increases. In other words, as far as the temperature coefficient of the resistance in the normal state is positive, ΔT is not affected by the strength of inter-grain coupling, implying absence of the paracoherent state. And the negative coefficient leads to the thermodynamic fluctuation of inter-grain coupling in the superconducting state, namely, the paracoherent state.

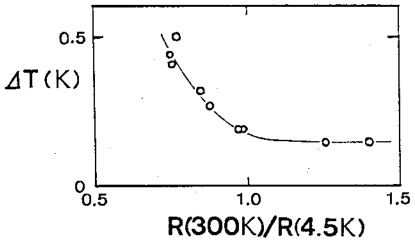

Fig. 6. Superconducting transition width vs. resistance ratio $R(300\ \mathrm{K})/R(4.5\ \mathrm{K})$. Transition width is defined as an interval between the temperatures giving 20% and 80% of $R(4.5\ \mathrm{K})$. Solid line guides the eyes.

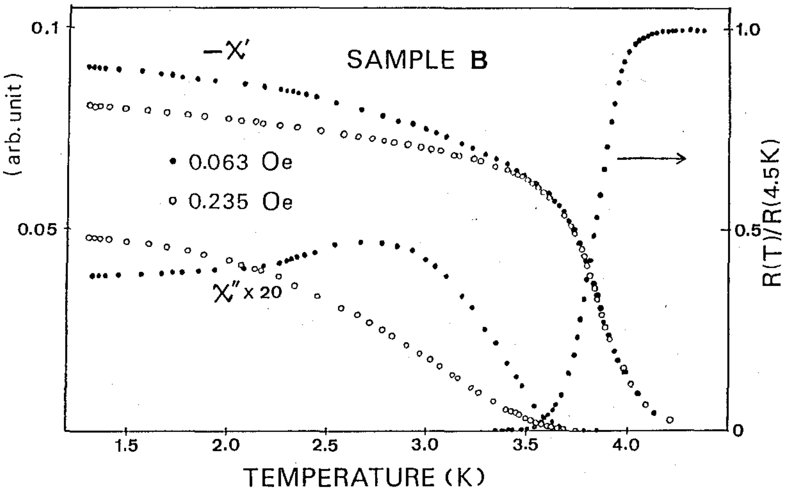

Fig. 7. Complex susceptibility vs. temperature on the sample B for different amplitude of external magnetic field, 0.063 and 0.235 Oe, 148 Hz. Normalized resistance is also shown.

2. Complex suceptibility

In Fig. 7, we show the complex susceptibility of the bulk granular system. The measurement was made on the sample B at different field amplitude, $h_0=0.063$, 0.235 Oe. As seen in the figure, we find:

(1) The onset of superconducting transition in χ' occurs at the same temperature as the resistive transition does.

(2) Above 3.7 K, χ' does not depend on h_0, while below that temperature, it shows h_0 dependence.

(3) χ'' appears only at 3.7 K and strongly depends on h_0. In particular, χ'' forms a peak against temperature for $h_0=0.235$ Oe.

First, according to the previously proposed model,[15] h_0-sensitive behavior of χ' and χ'' proves an appearance of diamagnetic current loops, through the inter-grain Josephson coupling. Besides this model predicts a peak formation in χ'' against temperature at small h_0. Experimental evidence below 3.7 K is the case in the present specimen. Second, there may be certain disorder in inter-grain coupling energies in the specimen. Such a quenched disorder characterizes superconducting transition as a percolating process under thermal fluctuation.

Taking into consideration above mentioned affairs, the observed features (1)–(3) are properly explained as follows: When the temperature decreases from 4.5 K, each grain becomes superconducting, and χ' increases from zero, reflecting the Meissner effect in each grain. Consequently, the system proceeds to achieve long range coherence between grains under thermal disturbance. However, due to disorder, superconducting grains preferentially begin to form clusters consisting of Josephson-coupled grains with higher than average coupling energy.[16] This percolating process causes the observed decreases in resistance. However, the clusters are not of massive closed loops down to 3.7 K, but of tying in a row with dead end arms. So not until at 3.7 K do closed loops begin to form and contribute to diamagnetic susceptibility as is seen in terms of h_0-dependence of χ' and χ''. Below 3.7 K clusters continue to merge and grow extending over the system.

Above explanation does not involve the thermal fluctuation of clusters. Recently a phenomenological description of inhomogeneous thermal transition has been proposed, where the percolation and the thermal transitions are together treated.$^{16)}$ Such a frame may be effective for analysis of the present experimental results.

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