

# Observation of a fluctuational tunneling conductivity of Cooper pairs in granulated superconductors

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“Zero-bias anomalies” have been detected in the differential resistance of granulated composites. These anomalies are caused by a fluctuational tunneling conductivity of Cooper pairs. The dependence of this component of the conductivity has been studied as a function of the temperature, a magnetic field, and an electromagnetic field.

In granulated superconductors with a Josephson interaction, the transition to a phase-coherent state near the metal-insulator phase transition is smeared out greatly along the temperature scale, a minimum is seen in the resistance, and reentrant effects are observed.<sup>1,2</sup> Several models have been proposed to describe the behavior of such superconductors. In the periodic model, for example, the suppression of superconductivity is explained by taking account of the short-range part of the electrostatic charge energy and the contribution of the Coulomb interaction of granules.<sup>3–5</sup> For large-grain granulated superconductors, the order parameter of the individual granules remain unperturbed, while the loss of long-range phase coherence in the case of the semiconductor model<sup>4</sup> and the precolation model<sup>6</sup> is attributed in large degree to a weakening to a Josephson link as a result of a decrease in the barrier transmission of grain boundaries. In two-dimensional granulated films, reentrant phenomena may result from an effect of quantum fluctuations on the dissociation of thermal vortex pairs.<sup>7</sup> Analysis of the existing models emphasizes the complexity of the problem of a coherent-phase state in three-dimensional granulated superconductors.<sup>8</sup> There is accordingly an extremely urgent need for experiments designed to identify the conductivity mechanisms in granular superconductors.

In the present experiments we have studied  $\text{BaPb}_{1-x}\text{Bi}_x\text{O}_3$  (BPB) composites with the perovskite structure. In these materials we have previously observed effects of reentrant phenomena on the temperature dependence of the critical current and the resistance. In the differential resistance of these materials, we found narrow “zero-bias anomalies” of the conductivity, due to the particular nature of a granular superconductor.<sup>2</sup>

The test samples are large-grain bulk samples of BPB with  $x = 0.2–0.3$ ,  $d_{\text{gr}} \cong 1–10 \mu\text{m}$ , and  $l \cong 10 \times 1 \times 1 \text{ mm}$ , synthesized by solid-phase diffusion from  $\text{BaPbO}_3$  and  $\text{BaBiO}_3$ .<sup>1)</sup> To accelerate the diffusion processes which cause the surfaces of the granules to become insulators, we held the samples, after synthesis, in a vacuum of  $10^{-5}$  torr. Such BPB samples have a semiconductor behavior over the entire temperature range (down to  $T = 1.5 \text{ K}$ ). Measurements of the susceptibility reveal a complete superconducting transition with  $T_{\text{c}}^{\chi} = 6–10 \text{ K}$ . In the experiments we study the dependence of the differential resistance on the temperature, a magnetic field, and an

electromagnetic field,  $dU/dJ(U)|_{T,H,P_{LO}}$ . From the characteristics  $dU/dJ(U)$  we single out a background hopping component  $\sigma_{hop}$  and a fluctuational component  $\sigma_f$  of the conductivity of the BPB (Fig. 1). The value of  $R_{hop}(0)$ , due to the intergrain resistance, was found through a quadratic approximation of the background part of the  $dU/dJ(U)$  curve from the region of high voltages in the anomaly region. The fluctuational component  $\sigma_f(0)$  was defined as  $\sigma_f(0) = \sigma_{samp} - \sigma_{hop}(0)$ . These measurements were carried out with several samples with  $\rho_{10K} = 1-100 \Omega \cdot \text{cm}$  in a cryostat with magnetic shields of the permalloy type, which cancel the stray pickup to a level of  $10^{-2}$  Oe. To record curves of  $dU/dJ(U)$ , we use the modulation technique which has become quite common in tunneling experiments.<sup>9</sup>

Figure 1 shows some typical curves of  $\sigma_f(0)$  as a function of the temperature ( $T$ ), a magnetic field ( $H_{||}$ ), and an electromagnetic field ( $P_{LO}$ ) for a BPB sample ( $x = 0.2$ ) with  $\rho_{10K} = 18 \Omega \cdot \text{cm}$ . For this sample, there is a sharp increase in  $R_{camp}(T)$  below the temperature of the superconducting transition of the grains ( $T_k^x = 6 \text{ K}$ ; see the inset in Fig. 1a). The "zero-bias anomaly" in  $dU/dJ(U)$  appears at  $T_k$ ,

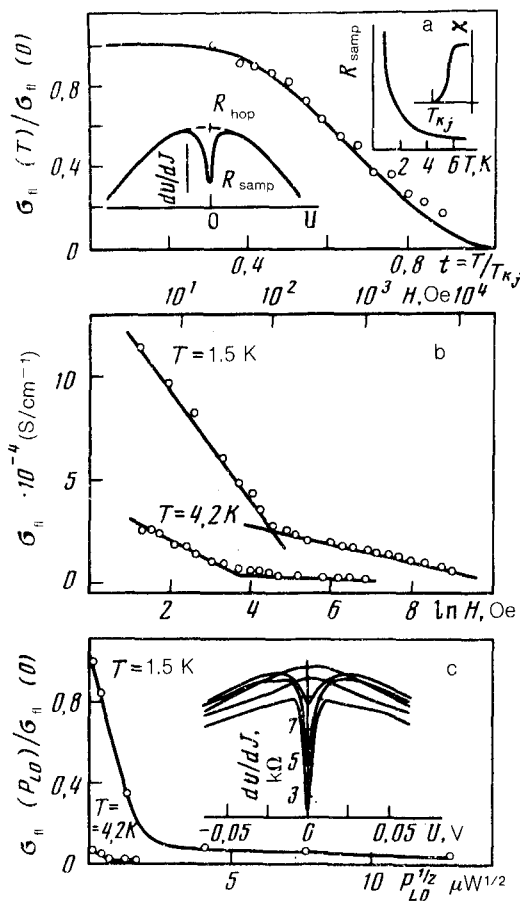


FIG. 1. Fluctuational component of the conductivity of the BaPb<sub>0.8</sub>Bi<sub>0.2</sub>O<sub>3</sub> composite as a function of the temperature, a magnetic field, and an electromagnetic field. The experimental values of  $\sigma_f(T)$  are shown in comparison with the theoretical prediction of the percolation model of a granular superconductor.<sup>6</sup> The insets show the temperature dependence of the total resistance and susceptibility of the sample (at the right in Fig. 1a), the typical curve of the differential resistance below  $T_k$  (at the left in Fig. 1a), and the effect of a microwave field on it (Fig. 1c).

$\cong 5$  K. This temperature corresponds to the nucleation of Josephson intergrain links in the ceramic. The behavior  $\sigma_n(T)$  is described by

$$\sigma_n(T) \sim \left[ \frac{\Delta(T)}{k_B T} \operatorname{th} \left( \frac{\Delta(T)}{2k_B T} \right) \right]^2,$$

found in the percolation model<sup>6</sup> for thermally excited tunneling Cooper pairs in a granular superconductor. Effects of the charge energy of the grains are unimportant here because of the large size of the granules.

A magnetic field suppresses the zero-bias anomaly in  $dU/dJ(U)$ . The  $\sigma_n(H)$  curve is similar to the  $J_c(H)$  curve for BPB composites, in which the superconducting current is not suppressed by a hopping component of the resistance of the surface layer of the granules. On the curve of  $\sigma_n(H)$  we can distinguish two regions (Fig. 1b). In the first region, the rapid changes in  $\sigma_n \sim 1/H$  ( $H < 200$  Oe) are caused by the loss of the phase coherence of the Josephson links at fragments of the percolation cluster for which the magnetic field vector lies in the plane of the intergrain tunnel junctions. In the second region ( $H > 200$  Oe), the slight changes in the remaining part of  $\sigma_n(H)$  (these terminate in fields  $H_{c2}$  in the transition of the grains to the normal state) can be attributed to the penetration of Abrikosov vortices into the matrix of granules or a suppression of the superconductivity of the remaining intergrain metal weak links. There is the further possibility that structural inhomogeneities are being manifested here.

A microwave electromagnetic field has its greatest effect on the superconducting anomalies in the resistance in BPB (Fig. 1c). Even a low radiation power ( $P_{LO} \cong 5 \mu\text{W}$ ,  $f = 2$  GHz) causes an essentially complete suppression of the zero-bias anomaly in  $dU/dJ(U)$ . These changes in  $\sigma_n(U)|_{P_{LO}}$  could not be due to a thermal effect of the field on the medium, since the background resistance  $R_{\text{hop}}$ , which is sensitive to the temperature, remains essentially constant in  $dU/dJ(U)$  at this microwave power. This component of the resistance decreases noticeably only at  $P_{LO} > 100 \mu\text{W}$ . The behavior  $\sigma_n(P_{LO}^{1/2})$  is similar to the behavior  $\sigma_n(H)$ . Its rate of change at low power levels is  $dR_n/dP_{LO} \cong 14 \times 10^9 \Omega/\text{W}$ . These dynamic processes in the conductivity of BPB during the imposition of microwave fields may be of a Josephson nature.

In the percolation model for a three-dimensional Josephson medium,<sup>6</sup> all of these results would be explained in terms of a fluctuational superconducting component of the conductivity. In the porous ceramic BPB, local variations in the potential barriers between granules give rise to a nonuniform distribution of the strength of the intergrain links, with the result that the static coherent-phase state is established in separate regions of the conducting path in the sample. A Josephson weak link is easily destroyed by transport current, with the result that the resistance of the percolation cluster increases. This process leads to the zero-bias anomaly in the conductivity. A characteristic feature of BPB composites is an additional disordering mechanism which results from the formation of an amorphous semiconducting layer at the surface of the granules, which leads to an electron localization. Here one observes, in contradiction of the conclusions of Ref. 10, a transition for percolation effects to a localization even in granular materials with large grains. This dominant localization effect

leads to a weakening of the fluctuational component of the conductivity as the temperature is lowered in BPB with  $\rho_{10\text{ K}} > 100 \Omega \cdot \text{cm}$  (Ref. 2). We believe that the behavior of BPB in magnetic and microwave fields is a consequence of the suppression of a coherent-phase state of fragments of an infinite cluster and a possible transition of the system into a spin-glass state. The transition to the state occurs in fields corresponding to the quantum of flux through a closed intergrain circuit with dimensions of  $0.1 \mu\text{m}^2$ , in agreement with electron-microscopy data.

We wish to stress that the modulation technique is extremely effective for studying the conductivity of granular superconductors. The experiments have revealed new aspects of the coherent-phase state of high-resistance  $\text{BaPb}_{1-x}\text{Bi}_x\text{O}_3$  composites and have revealed that they are highly sensitive to external electromagnetic radiation.

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