Quasireentrant superconducting transition in metastable states of a Zn–Sb alloy

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A quasireentrant superconducting transition in high-resistance states of a Zn–Sb alloy has been studied as a function of the transport current. The current–voltage characteristics of the test sample have voltage steps and a hysteresis. The maximum Josephson current decreases with decreasing temperature. There is a crossover from a Josephson characteristic to a tunneling characteristic when this current is exceeded.

Lin *et al.*[1] were the first to describe a reentrant superconducting (s) transition in a granular metal. The onset of a resistance and an increase in this resistance at temperatures below the transition temperature T_c have been observed in a Ba–Pb–Bi–O metal-oxide superconductor. The maximum value of the dissipationless Josephson current $I_c=I_c(T)$ on the current–voltage characteristics of the macroscopic test sample decreased with decreasing temperature in the range $T\leqslant(0.5–0.6)T_c$.

This behavior of the material was explained in Ref. 1 under the assumption that a tunneling between s-grains occurs through a degenerate semiconductor with a degeneracy temperature comparable to T_c. This effect could lead to a nonmonotonic $I_c(T)$ curve.[2] A similar behavior was later observed[3] in a K–Ba–Bi–O superconductor with a T_c three times as high. In the present letter we describe a similar behavior of a material with a very different composition: the metastable high-pressure s-phase of a Zn–Sb alloy, which undergoes a transition from a metallic phase to an insulating phase as a result of heat treatment.[4,5] The similarity in the behavior of these materials, with their very different T_c's, and with the different chemical compositions of the layers between the s-grains, is symptomatic. It indicates that the observed evolution of the s-response is governed by the structure of the granular materials, rather than being the result of a random coincidence of certain numerical parameters.

The Zn–Sb system is one of a group of systems in which there is a metallic phase at high pressure and high temperature, in which this phase can be quenched and preserved in a metastable state for an unbounded time at liquid-nitrogen temperature, and in which this phase is transformed by heating into an x-ray-amorphous insulating state. The latter state is also metastable, but there exists a certain temperature interval, near room temperature, in which this state persists for a fairly long time. If the composition is chosen correctly, the disordered state is uniform, i.e., does not have inclusions of other phases. Both the composition $Zn_{43}Sb_{57}$, with which we worked in Ref. 4, and the composition $Zn_{41}Sb_{59}$, which we studied in the present experiments, have this property.

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A corresponding property is exhibited by a Ga–Sb alloy, whose *s*-transition we have studied previously.[6] The alloys of both systems (Zn–Sb and Ga–Sb) undergo an increase in specific volume of about 25% upon the conversion to an amorphous state.[5] This factor promotes the formation of similar fractal structures in the course of the transformation.[7]

A pellet of the alloy $Zn_{41}Sb_{59}$, 5 mm in diameter and about 1.5 mm thick, which had been prepared previously, was held in a pressure chamber at a pressure of 70 kbar and a temperature of 350 °C for about 2 h. A transition to a high-pressure phase occurred in the solid state in the course of this treatment. The temperature was then lowered to liquid-nitrogen temperature at an average rate of about 20 deg/s. Then the pressure was removed. According to Debye patterns, the result was a polycrystalline sample with an orthorhombically distorted hexagonal structure.

At liquid-nitrogen temperature the pellet was placed in a holder, using two pairs of gold wires 0.5 mm in diameter with pointed ends to secure it. These wires doubled as electrical contacts. The holder with the test sample was placed in a cryostat, in which the temperature could be varied from 1.2 to 300 K. The resistance measurements were carried out in the current-source regime.

We were interested in a set of intermediate states between the original metastable metallic phase and the disordered insulating phase. A transition from one intermediate state to another was achieved by heating the sample to 155–160 K and by holding it there for 5–10 min. In the process, we continuously measured the resistance and found that it progressively increased. This hold was interrupted at a point such that the resistance of the test sample at the low temperature increased by a factor of 2 or 3. The state of the sample was not affected by any program of temperature changes below 120 K.

We characterize the instantaneous state of the sample by the parameter q, which is defined in terms of the resistance of the sample at $T=8$ K:

$$q=\log(R/R_{in})_{T=8\mathrm{K}}, \qquad (1)$$

where R_{in} is the resistance in the initial state, for which we have $q=0$. The value of R_{in} was 0.25 mΩ and corresponded to a resistivity on the order of 20–30 $\mu\Omega\cdot$cm. We selected 8 K as the temperature for this normalization. This temperature was dictated by the temperature of the *s*-transition in the initial state: $T_c(q=0)\approx 6.8$ K.

The evolution of the *s*-response in the Zn–Sb alloy is very similar to that which was observed[6] previously in Ga–Sb under similar conditions. In Ref. 6, however, the focus was on states with the highest values of q. At those values only the anomalous component of the *s*-response remained; i.e., the resistance did not decrease below T_c, and instead increased. In the present letter we are concerned primarily with intermediate states in which the dc *s*-transition has a typical quasireentrant shape.

Figure 1 shows curves of $R(T)$ in the $q=6.5$ state, recorded at various measurement currents (here R is to be understood as the ratio of the voltage across the potential contacts, U, to the measurement current I). The shape of the $R(T)$ curve depends on the measurement current. As this current is raised, the depth of the minimum decreases. At an even higher current, the minimum disappears completely,

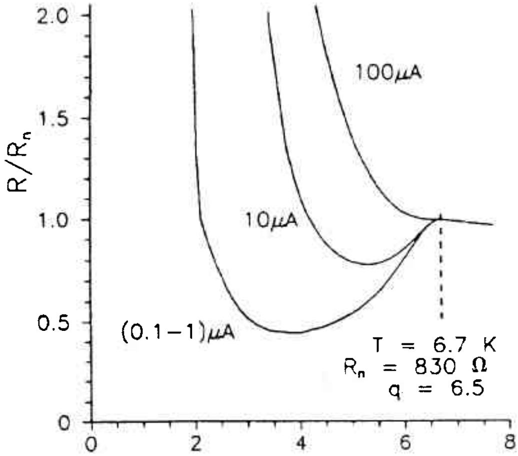

FIG. 1. Curves of $R(T)$ for the $q=6.5$ state for various measurement currents.

and the resistance increases with decreasing temperature everywhere below the transition temperature T_c. The depth of the minimum at a low current depends to some extent on the level of stray pickup and of random alternating signals in the electric circuits feeding the test sample. However, the resistance cannot be reduced to zero in this state. It is for this reason that this transition is called "quasireentrant."[8,9]

To construct a very simple model of a quasireentrant transition, we consider a 3D lattice of s-grains separated by intervals of a nonsuperconducting material. If the normal resistances r_n of these intervals have some scatter, the Josephson current in a fraction α of the junctions may be suppressed because of quantum fluctuations.[10] In this case, only a tunneling current of quasiparticles will flow across these junctions. This tunneling current will be exponentially small because of the presence of the s-gap Δ:

$$i \simeq (U/r_n)(\Delta/T)\exp(-\Delta/T). \tag{2}$$

If the fraction $1-\alpha$ is insufficient for the onset of an infinite s-cluster, the resistance of the sample, R, will decrease at T_c, but not all the way to zero. As the temperature is lowered further, the resistance R will begin to rise, because of the exponential increase in the tunneling resistance in accordance with (2). The s-response of our granular material is thus formed as the result of a competition between two components of this response:[11] a Josephson component, which is created by the s-currents between the s-grains, and a tunneling component, which is determined primarily by the s-gap Δ.

Figure 2 shows current–voltage characteristics of the sample at two temperatures. We wish to call attention to three features of these characteristics. First, there are a hysteresis and voltage steps as the current is raised. These features appear to be related to the presence of a multitude of parallel Josephson circuits and a redistribution of the currents upon the saturation of certain channels. Such steps have been observed in (for example) artificial reduced-dimensionality networks of Josephson junctions.[12,13]

A second feature of the curves in Fig. 2 is that the critical current I_c, which destroys the Josephson component of the s-response, decreases with the temperature.

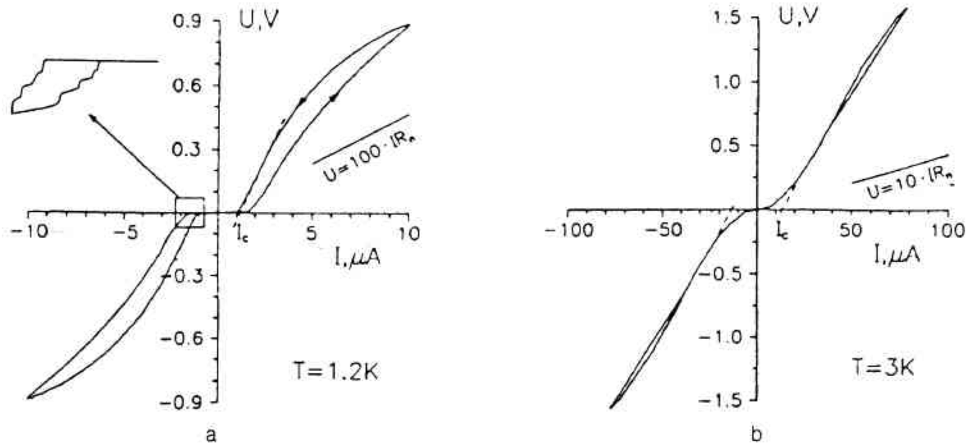

FIG. 2. Current–voltage characteristics of a test sample in the $q=6.2$ state at two temperatures. Normal characteristics $U=IR_n$ are shown for comparison (see the text proper for an explanation). The inset in Fig. 2a shows the first part of the loop in larger scale (in particular, the vertical scale has been increased by a factor of 10).

There is of course some arbitrariness in the method for defining I_c; our method is illustrated by the dashed lines in Fig. 2. Still, this arbitrariness could have only a slight effect on the overall shape of the $I_c(T)$ curve, on whose low-temperature branch I_c decreases by nearly two orders of magnitude (Fig. 3). In an isolated junction, the normal resistance r_n is related to the maximum Josephson current i_c by

$$r_n i_c(T)=\frac{\pi\Delta(T)}{2e}\tanh[\Delta(T)/2T], \quad r_n i_c(0)=\pi\Delta(0)/2e. \tag{3}$$

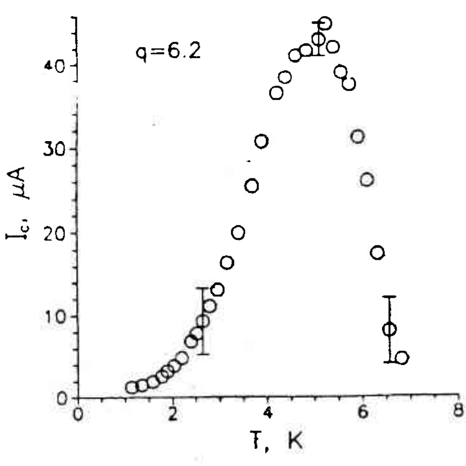

FIG. 3. Maximum Josephson current through a sample in the $q=6.2$ state versus the temperature.

It is usually assumed that r_n is independent of the temperature. This is a natural assumption if the conduction at the junction occurs as a direct tunneling between banks. However, there may also be *s*-junctions through semiconductor interlayers[2] or through an insulator with localized resonant centers.[14,15] It would then be more logical to assume that r_n increases with decreasing temperature. This behavior might affect the shape of the $i_c(T)$ curve.[2] This question has received little experimental study. However, the maximum on the $i_c(T)$ curve has also been seen in the case of single junctions, e.g., in the case of Sn–SnO–Sn structures.[16] We would also like to mention the experimental study by Roshchin *et al.*,[17] in which a maximum on an $i_c(T)$ curve measured at an isolated grain boundary in a Ba–K–Bi–O bicrystal was reported.

The third feature of the curves can be seen from a comparison of the *s*-characteristics, which we are discussing here, with the characteristics of normal junctions, $U=IR_n$, determined by extrapolating the temperature dependence $R_n(T)$ out of the normal region ($R_n=380\ \Omega$ at $T=8$ K, 425 Ω at 3 K, and 470 Ω at 1.2 K). In order to show the *s*-characteristics and the normal characteristics simultaneously in Figs. 2a and 2b, we were obliged to increase the ordinate scale for the latter characteristics, by factors of 100 and 10, respectively. These factors correspond in order of magnitude to the values of the function $\exp(\Delta/T)$ in Eq. (2), since the size of the gap Δ in our material is on the order of 10 K. This result means that when the Josephson junctions are destroyed by the current, the system goes into a tunneling-characteristic regime. This crossover from a Josephson characteristic to a tunneling characteristic has been observed not only in granular media,[1,3] but also at isolated junctions.[18] A necessary condition for this crossover is that the current i_c be much smaller than the value determined from (3):

$$i_c \ll \pi\Delta/2er_n. \tag{4}$$

It is likely that inequality (4) holds in junctions which are an intermediate case between high-resistance junctions with $r_n \approx \hbar/e^2$, for which we have[10] $i_c=0$, and low-resistance junctions, for which Eq. (3) is valid.

In summary, a quasireentrant *s*-transition has been observed in the *s*-medium in a 3D system of tunnel junctions, some of which exhibit Josephson properties. In this system we have also observed a hysteresis and voltage steps on the current–voltage characteristics, a decrease in the maximum Josephson current with decreasing temperature, and a crossover from a Josephson characteristic to a tunneling characteristic.

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