Negative Magnetoresistance in Homogeneous Amorphous Superconducting Pb Wires

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We report systematic magnetotransport measurements on a series of *in situ* grown homogeneous amorphous Pb wires. Corresponding to the broadening of the superconducting transitions as the wires cross over from 2D to 1D, we observe a *negative* magnetoresistance (MR) below the mean-field T_c at low fields. Both the magnitude of the negative MR and the crossover field increase with decreasing T_c and wire cross-sectional area. We speculate that these observations are the result of superfluid density fluctuations in sign as well as in magnitude along these wires close to the superconductor-insulator transition. [S0031-9007(96)02286-7]

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The nature of superconducting fluctuations in one dimension (1D) is concerned with the fundamental question of whether a true superconducting state exists in 1D. The study dates back to the 1960s, when Little [1] pointed out that thermal fluctuations could cause dissipation and excess resistance below the mean-field T_c in superconducting wires narrower than the superconducting coherence length. Subsequently, Langer, Ambegaokar, McCumber, and Halperin (LAMH) [2] worked out a detailed theory for the excess resistance below T_c based on a model of thermally activated phase slips. The excess resistance was predicted to follow a thermally activated behavior with an activation energy proportional to the superconducting condensation energy per unit volume. The theoretical prediction is in good agreement with the experimental data by Newbower *et al.* [3] for some cylindrical Sn whiskers about 0.5 μm in diameter. In the case of thermal fluctuations a zero resistance state can be reached even at finite temperatures. The thermal broadening of the superconducting transition in these wires is typically on the order of a few mK. With the advent of modern lithographic technologies it has become possible to fabricate wires with a much smaller cross section, making it possible to observe the effects of thermally activated phase slips over a significantly wider temperature range. As the wires are made narrower some qualitatively different features emerged, corresponding to deviations from the LAMH behavior. In a series of experiments, Giordano [4] observed a crossover from the LAMH behavior to a much more weakly temperature dependent excess resistance and the absence of a zero resistance state at $T = 0$, which he ascribed to quantum phase slips (macroscopic quantum tunneling of the phase). The interpretation of these results, however, was complicated by the possible granularity in the wires. It has been argued that the long resistance tails were not due to quantum phase slips, but rather due to weak links in the superconducting wires [5]. Indeed, in a set of experiments on *in situ* grown *homogeneous* Pb wires, Sharifi *et al.* [6] did not see these long resistance tails in wires as narrow as 150 Å and as thin as 10 Å. Instead, they found a less dramatic but systematic broadening of the superconducting transition beyond the LAMH limit as the wire cross-sectional area was reduced. On the other hand, long resistance tails are always present in intentionally fabricated granular wires [7].

In this Letter we present results of magnetotransport measurements on a series of homogeneous amorphous Pb wires similar to those in Ref. [6]. In these narrow wires we observe a *negative* magnetoresistance (MR) below T_c within the transition region, where broadening due to enhanced superconducting fluctions is observed in zero field [6]. The negative MR corresponds to a sharpening of the resistive transitions in a magnetic field. The observation of zero resistance and the negative MR provides strong evidence that the excess resistance observed in these wires is not the result of quantum phase slips.

We first fabricate a metallic stencil structure with a large overhang on a SiO_2/Si substrate. The substrate is then mounted inside a cryogenic evaporator, and the metal stencil serves as a shadow mask for the growth and *in situ* measurements of the wire. The apparatus is immersed in liquid He so that all the evaporations and measurements are carried out in UHV at low temperatures. Hence, we are able to measure a series of wires of the same width and different thicknesses in the same cooldown, avoiding sample to sample variations. The thickness of the wire is monitored with a quartz oscillator during deposition. We begin by depositing ~3 monolayers of Sb as a buffer layer before the Pb evaporations. It has been demonstrated that monolayer thick electrically continuous Pb films can be grown in this fashion [8], and that the films are homogeneous and not granular in morphology. The wire resistance is usually determined from the slope of the measured I-V curve at zero bias, and in some cases from direct low-frequency phase sensitive measurements. In the case of ac measurements, the current is kept below 10 nA to ensure that the measurements are in the Ohmic regime, and dc I-V curves are taken at different points throughout the experiment to ensure that the results are consistent. Each wire consists of three sections of different lengths, 2, 1.5, and 1 μm, so that we can check the consistency and the length dependence of our results. In all of the samples we

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measured, the wire resistance scales with the wire length above T_c.

Figure 1(a) shows a typical set of $R_\square$ vs T data in zero field for these uniform Pb wires. The wire width here is 350 Å and the variable is the thickness. The superconductor-insulator transition (SIT) looks qualitatively similar to that in 2D. Nevertheless, there are many signatures of a dimensional crossover to 1D, both in terms of superconductivity and electron transport in the insulating state. Note that we continue to use the 2D parameter $R_\square$ for the purpose of comparing the behavioral differences exclusively due to variations in the wire width. In the insulating state, the wire resistance exhibits a much stronger T dependence than that of 2D films [9], indicating that 1D Coulomb correlation effects play an important role in the transport properties of the wire. On the superconducting side, we see a consistent suppression of T_c as the wire width decreases, as shown in Fig. 1(b). Most likely, this is also the result of the enhanced Coulomb interaction. Superconducting fluctuations, on the other hand, cross over into 1D at a relatively large length scale because of the divergence of ξ at T_c. The wires studied here are in the 1D limit as far as superconducting fluctuations are concerned, as evidenced by the fact that the widths of the resistive transitions scale with the wire resistance rather than $R_\square$ [6]. The T_c is determined by fitting the upper half of the resistive transition to the Aslamasov-Larkin model [10], and the result is close to the temperature at the transition midpoint $R_N/2$. There is no longer a true phase transition at this temperature because of the fluctuations. Yet this temperature still signifies a point below which there is a substantial superconducting order on the wire, although long range phase coherence is not established until a lower temperature.

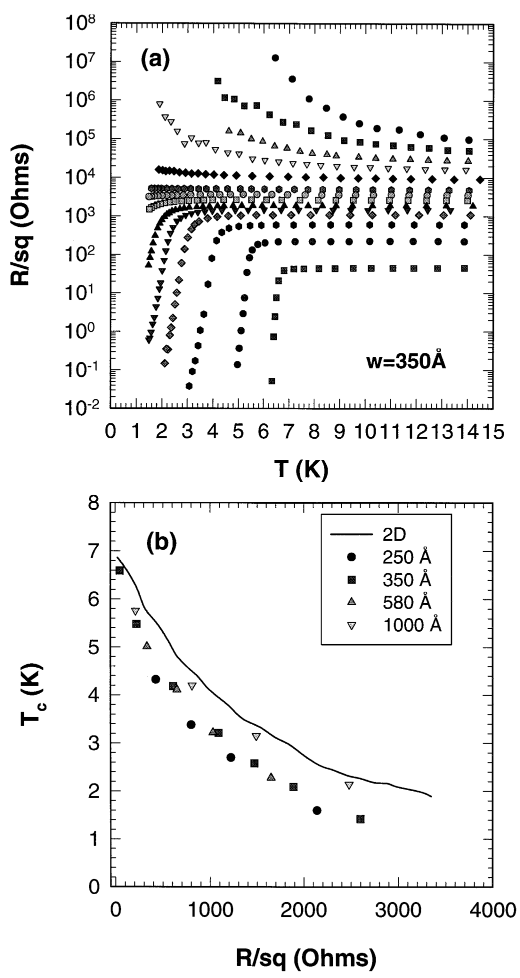

FIG. 1. (a) $R_\square$ as a function of temperature for a 350 Å wide wire at different thicknesses. (b) T_c as a function of the normal state sheet resistance for a 2D Pb film and four wires of different widths.

The application of an external magnetic field would be expected to enhance superconducting fluctuations. The magnetic field raises the energy of the superconducting state, thereby reducing the energy barrier for phase slips and broadening the resistive transitions. To our surprise, we observe that a small magnetic field actually suppresses superconducting fluctuations and sharpens the resistive transitions in 1D wires. The wires exhibit a pronounced negative MR in the transition region. In our experiments, the field is always perpendicular to the film plane. Figure 2(a) shows the zero-field transport curves for a 520 Å wide wire. The symbols on the curves indicate the points where MR measurements were made at a constant temperature. Three field sweeps at point α are displayed in Fig. 2(b). The MR does not show any hysteresis, and it does not depend on the field direction. The wire resistance decreases by more than a factor of 5 at $H = 2.5$ kOe before the pair-breaking effect takes over and the resistance increases. In Fig. 2(c) we attempt to compare the effect at different thicknesses (T_c's). Previously this comparison was extremely difficult, and it is now made possible by our ability to change the wire cross-sectional area *in situ.* Still, the comparison is not quantitative, because the negative MR depends sensitively on the exact position in a transition, as shown in Fig. 3. Nonetheless, the qualitative trend is clearly demonstrated; the effect becomes stronger as T_c is lowered, both in terms of the magnitude of the negative MR and the crossover field.

Figure 3 demonstrates the sensitive temperature dependence of the negative MR. Here we compare the effect at different positions along the same transition, as indicated by the symbols in Fig. 3(a). From Figs. 3(b)–3(e), it is obvious that the negative MR is most pronounced near the end of the resistive transition, and it diminishes quickly moving up in temperature and closer to T_c.

We now look at the effect of the wire width. Again we want to emphasize that it is difficult to precisely compare the negative MR in wires of different widths because of the sensitive T and T_c dependence just described. Figures 4(a)–4(d) show $R_\square$ vs H for four wires of different widths ranging from 250 to 1000 Å. In choosing these points, we have attempted to pick a point at the same position along a transition with the same T_c for each wire. Despite the aforementioned complications, the strong depen-

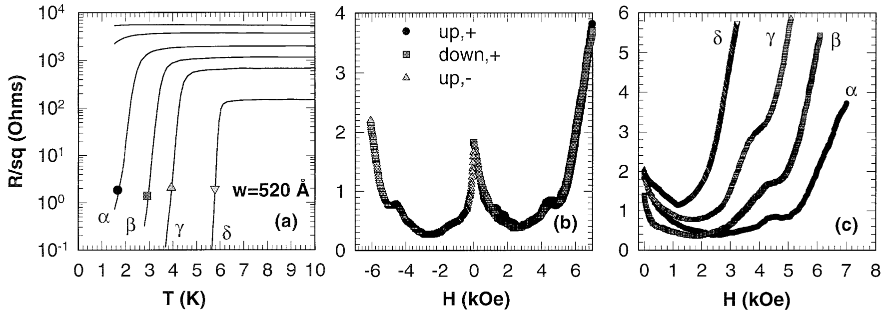

FIG. 2. (a) Resistive transitions for a 520 Å wide wire. The symbols represent the points at which we measured the magnetoresistance. (b) Three field sweeps at point α. (c) Magnetoresistance at points $\alpha - \delta$. The temperatures at the four points are 1.63, 2.90, 3.93, and 5.78 K, respectively.

dence of the negative MR on wire width is apparent; the negative MR increases with decreasing wire width.

The evolution of the negative MR, with wire cross-sectional area and temperature as described above follows the evolution of the zero-field excess broadening beyond that predicted by the LAMH theory. There appears to be a close correlation between the two effects. This correlation is further evidence that the excess broadening is not due to edge roughness of the wires, but instead is an intrinsic dimensional effect. It also suggests that the negative MR is a manifestation of the suppression of the superconducting fluctuations by the magnetic field. To summarize, we have observed an enhancement of superconductivity and a suppression of superconducting fluctuations by a perpendicular magnetic field in thin homogeneous Pb wires, and the effect is a result of the dimensional crossover from 2D to 1D.

Negative MR was observed close to T_c in superconducting Sn stripes [11] and Al loops [12]. The negative MR was attributed to the nonequilibrium charge imbalance process [13] at normal-superconducting (N-S) boundaries, produced either intrinsically by spatially localized phase-slip centers due to high current density [11] or artificially by ion damage [12]. The magnetic field reduces the charge imbalance relaxation time and hence

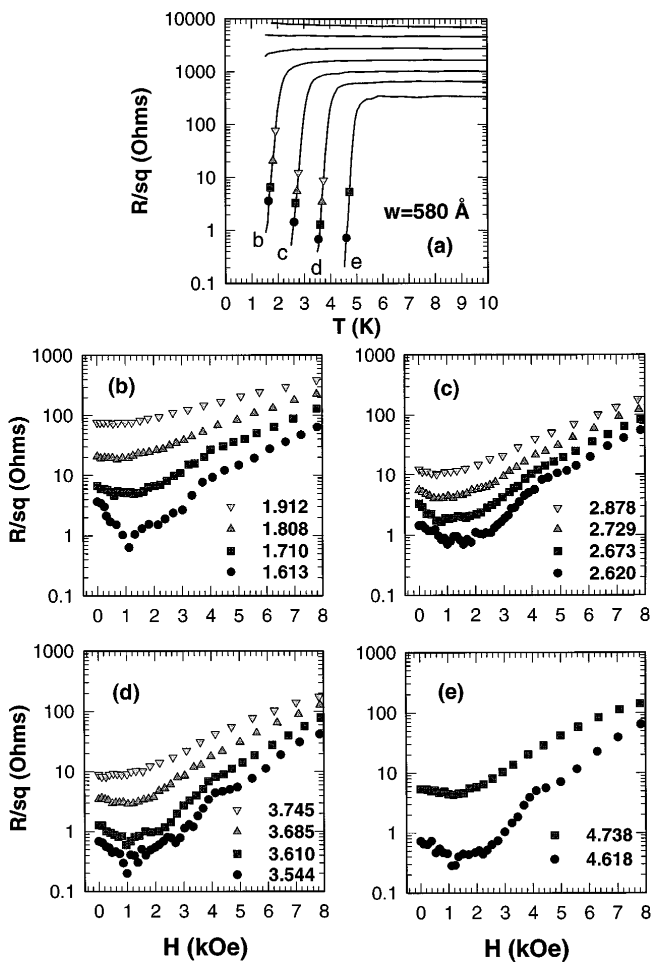

FIG. 3. (a) Resistive transitions for a 580 Å wide wire. The symbols represent the points at which we measure the magnetoresistance. (b)–(e) Magnetoresistance for the wire at different thicknesses. The numbers indicate the temperature in Kelvin.

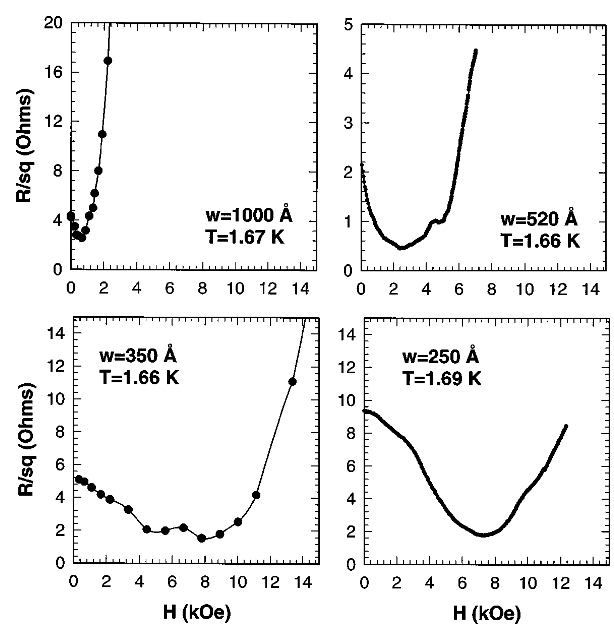

FIG. 4. Magnetoresistance for four wires of different widths: (a) 1000 Å; (b) 520 Å; (c) 350 Å; (d) 250 Å. The measurements were taken at approximately the same position along transitions of approximately the same T_c for each wire.

the N-S boundary resistance [11,12]. In our wires there are no long-lived localized phase-slip centers, and the negative MR is obtained in the zero-current limit. The relaxation time for thermally activated phase slips is too short for the charge imbalance process to have a significant effect. Therefore, we do not believe that this mechanism is responsible for our observations.

Another possible cause for the negative MR is the quenching of the pair-breaking effect of Kondo impurities by the magnetic field [14]. We know that there are a large number of localized states in the Sb buffer layer [15]. The Coulomb correlation could destroy the spin degeneracy of these states; i.e., they could be singly occupied. The single localized electrons could conceivably act as Kondo impurities and cause pair breaking. The application of a moderate magnetic field would quench the spin fluctuations and reduce the pair-breaking effect, thus raising T_c and producing a negative MR. However, we do not see an enhancement of T_c in field. The negative MR is observed only below T_c. Also, in our temperature range the Zeeman energy of the localized electrons in a kOe field is an order of magnitude smaller than the thermal energy. Namely, the magnetic field at which the negative MR is observed is too low to overcome the spin fluctuations. Furthermore, it is difficult in this model to explain the fact that the observed negative MR is a dimensional effect.

Several years ago a mechanism for negative MR was proposed by Kivelson and Spivak [16], specifically for highly disordered superconductors close to the SIT. In this picture, the negative MR originates from the possible existence of negative Josephson coupling between superconducting grains in granular films [17] and negative superfluid density in homogeneous films [18]. The local critical current I_c in these systems is determined by the interference between regions with opposite signs of superfluid density. When there is a substantial fraction of local regions with a negative superfluid density, the first-order interference term will be averaged out in the ensemble-averaged I_c, while the second-order term will have a negative sign [16]. Therefore, I_c increases for small fields, which gives rise to a negative MR in the low-field limit. We understand that the original theory was proposed for 2D uniform and granular films close to the SIT, and negative MR was not observed in either case [19,20]. We believe that in the granular films this implies that the intergrain Josephson coupling is dominated by direct pair tunneling, which always yields a positive coupling energy. In the uniform films there is evidence that there are strong fluctuations in the amplitude of the superconducting order parameter close to the SIT [19]. It is expected that 1D Coulomb correlations [21] should result in even stronger fluctuations in the superfluid density along the wire. It has been suggested that the superfluid density fluctuations are the origin of the excess broadening in the narrow uniform wires [6]. We speculate that 1D Coulomb correlations not only enhance fluctuations in the magnitude, but also induce fluctuations in the sign of the local superfluid densities. This could be the origin of the negative MR in our wires. In fact, in Fig. 4 the MR curves show clear evidence for the resistance oscillations expected from such interference loops [16]. The "oscillation" period increases with decreasing wire width and is approximately the size predicted by the theory.

Further work on well-defined loop geometries will enable us to elucidate the resistance oscillation period more accurately and provide a more definitive answer to the origin of the negative MR.

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