Disorder-induced Andreev reflections in granular metals

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We have studied the transport properties of SNS structures where N is a disordered Ag film quench condensed into a narrow gap between two superconducting Pb electrodes. This setup enables us to control the disorder of the normal region by sequentially depositing layers of Ag *in situ*. We find that when the length of the disordered N section is 1.5–2 μm the samples exhibit a subgap resistance minimum, which we interpret as evidence for strong Andreev reflection processes. This feature evolves into the usual Blonder-Tinkham-Klapwijk behavior as we reduce the disorder of the N region by increasing the Ag thickness. In samples which are shorter than 1 μm, this crossover is absent. We discuss the results within the framework of the ''reflectionless tunneling'' mechanism. [S0163-1829(99)06713-2]

Andreev reflections[1] are processes responsible for charge transfer through a contact between a normal metal (N) and a superconductor (S). An electron in the normal metal impinging on the interface with energy smaller than the superconducting gap Δ couples with its time-reversed electron, adding a Cooper pair to the superconductor. As a result, a hole is retroreflected from the interface. The effect of these processes on the *I-V* characteristics of S-N junctions was discussed in detail by Blonder, Tinkham, and Klapwijk (BTK).[2] When the transmission coefficient of the junction Γ is unity, every electron impinging on the interface undergoes an Andreev reflection and is accompanied by a retroreflected hole. Because the charge transfer associated with these processes is $2e$, the resistance at subgap voltages is half of that at $V > \Delta$, where Andreev reflections are not effective. In the other limit, when Γ is close to zero, the behavior is that of a tunnel junction and the resistance at subgap voltages is very large. The *I-V* curve of any intermediate case can be thought of as a superposition of these two extremes. Real samples do not generally exhibit perfect transmission even for high-quality contacts because the mismatch of the Fermi wave functions on both sides of the junction gives rise to an intrinsic nonzero reflection coefficient. Hence, a realistic resistance versus voltage (*R-V*) curve is characterized by a double-dip structure at $V = \pm\Delta$ and a peak centered at $V = 0$. BTK theory has been successful in providing a method to extract the junction transmission coefficient based on experimental *R-V* curves.

Over the past few years there have been a number of experiments on S-N contacts where N was a semiconductor[3] or a metal with a confined geometry,[4] which showed an anomalous resistance dip at zero bias. These results have been interpreted as enhancement of the Andreev probability due to the disorder in the normal region.[5] The enhancement is envisioned as taking place in the following way: In the mesoscopic limit, $L_e < L < L_\varphi$ (where L is the sample length, L_e is the elastic mean free path, and L_φ is the phase coherence length in N), a charge carrier will be scattered elastically many times before it loses phase memory. Under these conditions, an electron reflecting off the superconducting interface after failing an Andreev reflection, may be scattered coherently from the impurities in the normal material and impinge the interface again as illustrated schematically in Fig. 1. Hence, the electron has another ''chance'' at an Andreev process while retaining its coherence. This process, together with the phase conjugation between the electron and the reflected hole gives rise to an *enhancement* of the Andreev coefficient in the system. The mechanism responsible for this enhancement has been named ''reflectionless tunneling'' because the multiple scattering reduces the effect of the barrier and may increase the transmission by a large factor.

Because the reflectionless tunneling process relies upon back scattering in the N region, it requires a number of conditions. First, the disorder must be large enough to allow multiple returns to the interface. Hence, L_e should be relatively small. Second, the length of the normal region L has to be large enough so as not to allow the electron to escape the mesoscopic region before it undergoes an Andreev process. On the other hand, the mechanism depends on quantum coherence and therefore it is necessary to use samples in which L is not much larger than L_φ. If L were too large, the measured resistance would be completely dominated by the disordered region and the effect of the interface reflectivity

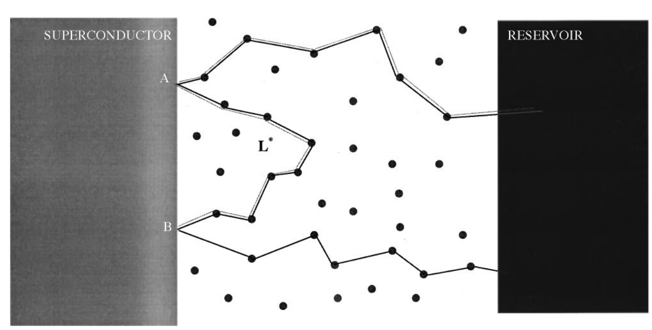

FIG. 1. A schematic description of a possible trajectory of a particle in the disordered N region. An electron (solid line) bouncing of the interface at point A follows trajectory L_* and hits the interface again at point B. Now it can either reflect normally and return to the reservoir as an electron or undergo Andreev reflection and return as a hole (dotted line). Positive interference of the electron-hole pair in points A and B increases the probability for the Andreev process. This mechanism is more effective the larger the number of times the electron returns coherently to the interface.

would not be measurable. These considerations set the following restriction on the relevant length scales in the system[5]

$$\frac{L_e}{\Gamma} \ll L < L_\varphi . \tag{1}$$

The interesting notion which emerges from this model and provides the motivation for the present work is that increasing either the disorder of a sample or its physical length can *increase* the excess conductance due to Andreev reflections. Thus, it is of great interest to study an S-N system in which the disorder of the N region and its length can be varied systematically. In this paper we present measurements on *I-V* characteristics of an SNS planar structure junction where the N is a disordered granular metal with dimensions comparable to or smaller than L_φ. We vary L, L_e, and L_φ in a controlled way and determine the influence of these changes on the electric properties. For samples of length 1.5–2 μm we observe a transition from anomalous Andreev reflection dominated behavior to conventional BTK type *I-V* curves as the disorder of the normal metal is decreased. The anomaly is not detected in samples shorter than 1 μm or when the temperature is raised so that $L > L_\varphi$.

We have chosen to fabricate the disordered metallic samples using the method of quench condensation (evaporation at cryogenic temperatures under ultrahigh vacuum). This technique enables us to vary the disorder of a specific sample by sequentially evaporating thin layers of material *in situ*. Transport measurements as well as scanning-tunneling microscopy images[6] show that at the first steps of deposition a typical quench-condensed metallic film contains isolated grains of 50–150 Å in size and the sample is insulating. Adding material smoothens the surface and couples the grains causing reduction of the resistivity and sample disorder. Thus, we can go through the metal-insulator transition and reach far into the metallic regime using a single sample. Furthermore, we have access to a metallic regime in which L_e is extremely small. Our Ag films have L_e of the order of a few tens of Å (for sheet resistances, $R_\square$, of a few tens of ohms) whereas in previously studied semiconductor systems the values reported for L_e were 500–1000 Å.[3] In the experiment reported here, the short mean free paths are apparently of relevance to the observed *I-V* characteristics as discussed below.

The samples were prepared in the following way: We began by thermally evaporating a strip of Pb, connected to four leads, on a room temperature Si substrate. We then cut a narrow slit in the Pb strip using e-beam lithography and dry-etching techniques. The width of the slit, which defines the length of the normal region of our SNS junctions, varied between 0.1 and 2.5 μm. In order to reduce the effect of a tunnel barrier at the interface we removed any oxidized Pb by plasma etching the surface of the lead layer and covering it with an ultrathin (20 Å) film of Ag *in situ*. However, even a perfect interface between two metals has a finite electron reflectivity due to the Fermi velocity mismatch at the boundary between different materials. Such a reflectance is an essential ingredient for the Andreev reflection enhancement mechanism. Next, we mounted the sample in a cryogenic evaporator and quench-condense sequential layers of Ag into the gap under UHV conditions up to a total thickness of 200–300 Å. We monitor the resistance and thickness of the sample during the deposition and measure the transport properties at desired resistances. Dynamic resistance versus voltage (dV/dI-V) measurements were performed using ac lock-in methods in a rf shielded room.

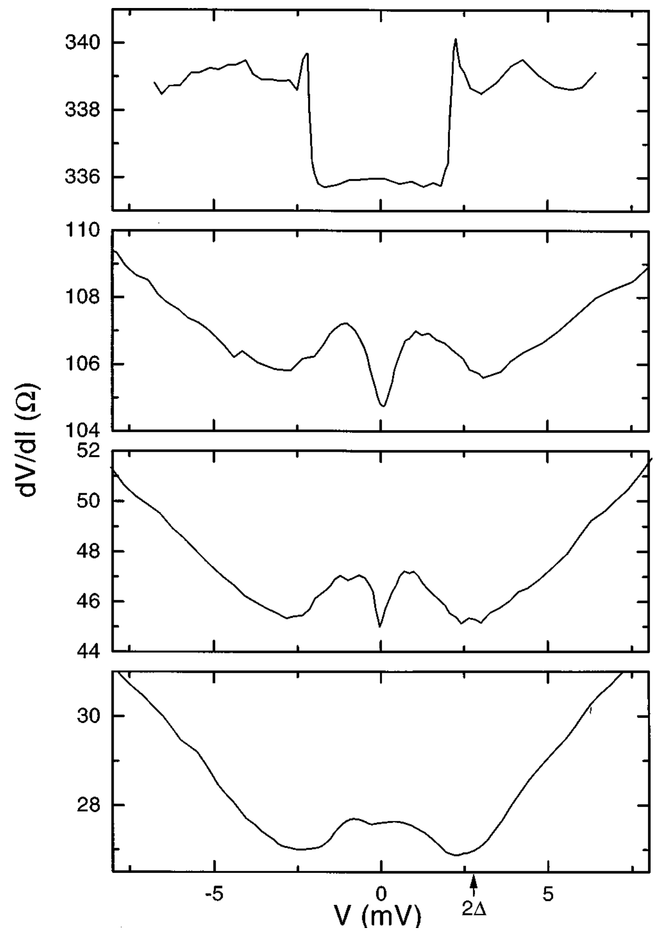

FIG. 2. Dynamic resistance versus voltage curve for a 2 μm sample at different evaporation stages. $T = 1.8$ K. The corresponding Ag thicknesses are 58, 69, 86, and 118 Å, respectively.

Figure 2 shows the evolution of the dynamic resistance of a sample of $L = 2$ μm for different resistances. It is seen that the dV/dI-V curve exhibits a transition from a wide resistance minimum at subgap voltages, through a narrower dip, to a usual BTK-like curve as the resistance is decreased. Since the samples consist of two S-N interfaces in series, the relevant features occur on a voltage scale of 2Δ, which, for Pb, is about 2.8 mV. It is significant, we believe, that the voltage scale over which these effects are observed is the energy gap of the Pb electrode. This suggests that the resistance changes are a result of interface effects. If the resistance were dominated by the bulk of the sample we would not see these effects at a voltage of 2Δ. Indeed, in the initial stages of the Ag quench condensation we observe features at voltages higher than 2Δ indicating that a significant part of the voltage drops on the N region. As more material is deposited and the interface dominates the resistance, the features in the resistance curves shift towards lower voltages and saturate at the gap value. At voltages larger than the gap there is an evident voltage-dependent background of the resistance, causing an increase of R with V. Though the origin

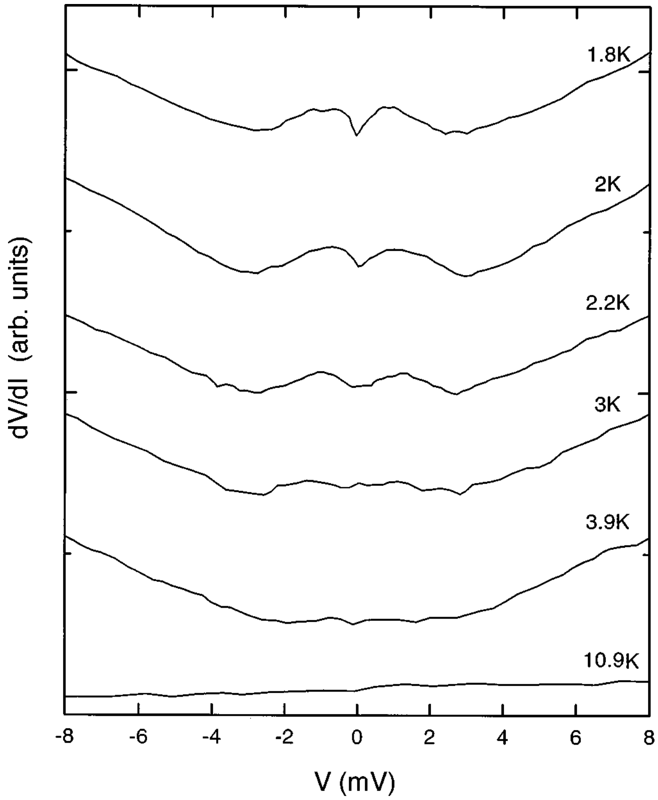

FIG. 3. dV/dI-V for the $R_{\square}=50\ \Omega$ sample presented in Fig. 2 at different temperatures.

of this non-Ohmic behavior is not entirely clear it seems reasonable to ascribe it to inelastic processes. L_{φ} is expected to be energy dependent and will decrease with increasing voltage thus causing a resistance increase.

The results presented in Fig. 2 demonstrate that *decreasing* the disorder of the N region causes a reduction in the subgap excess current consistent with the reflectionless tunneling mechanism. An important feature in the data is that for a large disorder the resistance minimum extends over a voltage range of the entire gap. In previously reported cases, the resistance dip was highly energy dependent and vanished for voltages much smaller than the gap [typically, the excess conductance was completely suppressed for $V>0.5$ mV (Refs. 3,4)]. This sensitivity to voltage is due to the fact that the electron-hole phase conjugation is maintained only in the absence of an electric and magnetic field, H. For $V,H>0$ the phase difference between the electron and the hole wave functions is given by[5]

$$\Delta\phi=\frac{2eVL^*}{hv_F}+\frac{HA}{\Phi_0}. \tag{2}$$

The second term arises from loss of phase coherence due to Aharonov-Bohm flux penetrating the area, A, enclosed by the trajectory L* between two intersecting points with the barrier (points A and B in Fig. 1) and the superconducting interface. The first term reflects the fact that the electron and the hole accumulate different phases while traveling in a trajectory of length L* in the presence of an electric field. Random-walk calculations and simulations show that the length of a typical trajectory between two points in which a diffusing particle hits the interface, L* (between points A and B in Fig. 1 for example), is proportional to L_e. Therefore, the characteristic voltage for the suppression of the electron-hole phase correlation is inversely proportional to the elastic mean free path. Since L_e in our samples is at least an order of magnitude smaller than that in previous studies, the voltage scale of enhanced Andreev process may be larger by a similar factor. This might be the reason for the relative robustness to applied voltage in our quench-condensed films. Evidently, our data show that apart from reducing the amplitude of the subgap resistance minimum, reducing disorder also increases the sensitivity to electric field as one might expect.

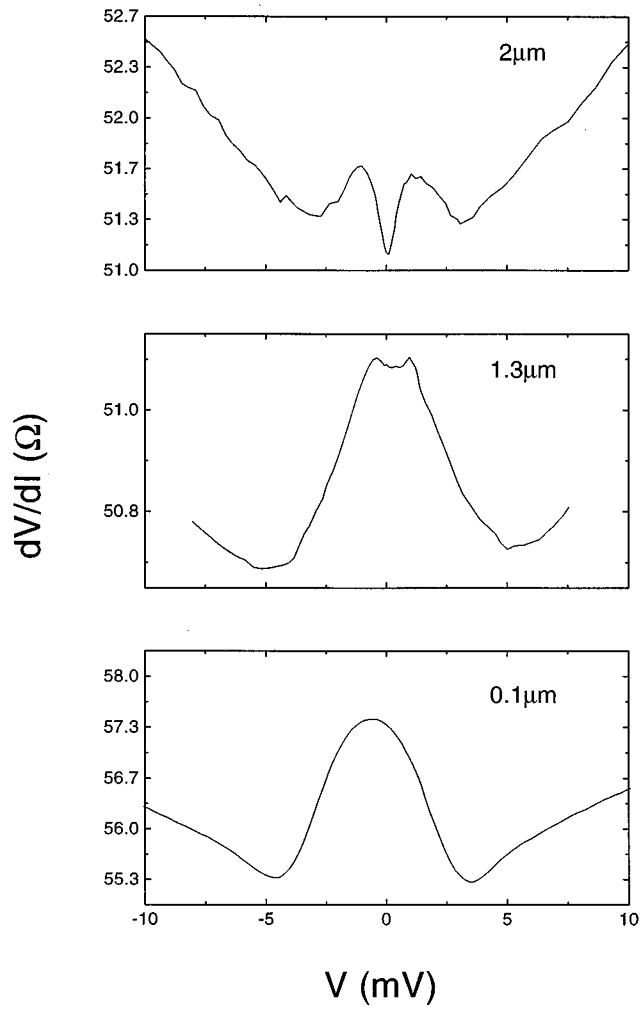

FIG. 4. Dynamic resistance curve of samples with different lengths having a sheet resistance of about 150 Ω. Note that for the shorter samples the resistance dip occurs at voltages slightly above 2Δ. This is typical of samples in the early evaporation stages due to the series resistance of the disordered metal. As more material is deposited the dip voltage shifts and saturates at 2.8 mV.

We now address the issue of quantum coherence. Weak localization measurements on quenched-condensed films of Ag similar to ours[7] having a sheet resistance of 100 Ω showed that the inelastic length is 0.5 μm at $T=4$ K and extrapolates to 1.5–2 μm at 1 K. Thus, it is reasonable that our 2 μm samples can barely support quantum coherent effects at 1.5 K. Figure 3 shows the temperature dependence of the dynamic resistance curve for the 50 Ω sample presented in Fig. 2. The subgap dip decays rapidly with increasing temperature and disappears at $T=3$ K. One might expect that reducing the size of the sample so that $L \ll L_\varphi$ would assist the Andreev reflection enhancement mechanism since quantum interference processes are more easily realized. This is not the case. Figure 4, which depicts dV/dI-V curves for samples with different L, shows that for a 1.3 μm long sample only a weak subgap resistance minimum is seen and when $L=1000$ Å there is no sign for a resistance dip at all. The absence of the dip is common to all samples we measured with lengths in the submicron regime. Clearly, the Andreev reflection enhancement decreases as the disordered N region becomes shorter. This result is also in qualitative agreement with the reflectionless tunneling mechanism [as presented in Eq. (1)]: The shorter the length of the normal region the larger the chance of an electron after impinging on the superconducting interface to ''leak-out'' to the other side without undergoing Andreev reflection. Once the electron reaches the opposite superconductor, the enhancement process is not effective since the particle can be transmitted into both superconductors with similar probabilities.

It is perhaps valuable to reiterate the physical picture we have for this conduction enhancement. One possible mechanism for such an effect is simply that observed in an SNS proximity junction (having clean S-N interfaces) due to an enhancement of the diffusion constant in the disordered region. This does not seem to be the case in our junctions. If it were, the effect would be even more pronounced for narrower structures (smaller L). The opposite is the case as the effect disappears for small L. The observation that the energy scale is always 2Δ (Δ for each interface) strongly suggests that the interface between the superconductor (Pb) and the metal (Ag) plays a dominant role. If the disordered region dominates the conduction, one would expect the relevant energy scale to be approximately the Thouless energy (which depends on the sample length and disorder), not the superconducting gap. The picture then is that a diffusing electron that is scattered at the Pb/Ag interface and not transmitted into a pair state, gets another chance before losing phase coherence in the highly disordered metal. If the interface is a significant part of the resistance this will result in a *reduced* resistance at subgap voltages.

As mentioned, the resistance minimum is not observed for short samples. Nevertheless, there might be evidence for the disorder-induced Andreev mechanism in the data of these samples as well. When examining dynamic resistance curves for samples with $L<1$ μm we notice that the relative magnitude of the zero-bias peak increases with decreasing resistance. We have not been able to achieve satisfactory fits to BTK theory[8] but we can estimate relative changes in junction transmission by studying the ratio between the amplitudes of the zero-bias peak and the dips at the gap voltage. For samples with L=1000 Å this ratio typically grows by a factor of 1.1–1.4 as $R_\square$ is decreased from 1000 to 40 Ω. We speculate that this may be a sign for a latent resistance dip which is overshadowed by the usual BTK peak. The importance of this dip becomes smaller as the sample resistance decreases. Thus, the reflectionless tunneling mechanism, though very weak in these short samples, still affects the transmission of the junction for large enough disorder and it becomes less effective with decreasing disorder.

In summary, we have studied electric transport properties of an interface between a superconductor and a disordered metal as a function of the disorder for different sample sizes. We show that decreasing the disorder in the normal metal leads to a reduction of the excess current associated with Andreev processes. Another impact of lowering the disorder is the reduction of the voltage scale over which the Andreev enhancement is effective. In addition, we see that as the junction length is made smaller, the Andreev processes become less important.

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