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Transition from the tunneling regime to point contact and proximity-induced Josephson effect in lead–normal-metal nanojunctions

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We have studied the transition from the tunneling regime to point contact with a low-temperature scanning tunneling microscope, when one of the electrodes is a normal metal (Au, Pt-Rh) and the other is a superconductor (Pb). Continuous variation of the junction resistance, spanning more than six orders of magnitude, results in the observation of the superconducting gap, Andreev reflection, and for low resistance, a zero-bias sharp peak. Detailed study of the conductance curves and comparison with results for junctions composed of two superconducting electrodes support the interpretation of this feature as a second-order proximity-induced Josephson effect (PJE).

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A point contact between a superconductor and a normal metal (SN contact) or between two superconductors (SS′ contact), is a convenient geometry to study different aspects of superconductivity. The possibility of varying the strength of the potential barrier and the contact area between the electrodes allows the observation of a large number of phenomena.

In the range of high tip-flat resistances, conduction between the electrodes is due to quasiparticle tunneling. The resulting conductance reflects the quasiparticle spectrum and is explained by the BCS model. It shows a dip around zero bias, since tunneling is not allowed for energies within the superconducting gap.

As the tunneling barrier collapses, low voltage conduction is governed by the Andreev reflection process. This transition from tunneling to contact has been studied theoretically and experimentally by Blonder, Tinkham, and Klapwijk (BTK),[1,2] within a generalized semiconductor model, using the Bogoliubov equations to treat the transmission and reflection of particles at the interface. In this case, although an incoming electron from the normal metal cannot continue as an electron in the superconducting metal if its energy is below Δ, the superconducting gap, it may be reflected as a hole in the normal metal while simultaneously adding a Cooper pair to the condensate in the superconducting metal. This process causes an increase in conductance around zero bias. If the tunneling barrier is wide the probability of the Andreev reflection process is negligible and the BTK model yields a conductance identical to the BCS model. But as the strength of the barrier diminishes, probability increases and conductance curves show a central hump. The maximum conductance at zero bias is obtained when tip and flat are in contact.

As is well known, Josephson effects are normally observed, in addition to quasiparticle tunneling, when both electrodes are superconducting and the barrier is relatively low. Similar effects have also been observed in SN contacts[3,4] with N a normal metal or superconductor above its transition temperature, and, more recently, in SSm contacts (where Sm is a semiconductor).[5] The initial interpretation of these phenomena[6] as Josephson effects between the weak superconductivity induced in the surface of the normal electrode by the proximity effect and the superconductor [proximity-induced Josephson effect (PJE)], has been questioned by Kadin.[7] This author argues that a standard first-order Josephson effect across an SN contact is fundamentally impossible and proposes an alternative that requires the existence of a phase-slip center (PSC) in the superconductor near the SN interface. As a consequence the observed Josephson effects would be taking place in the superconductor. It must be remarked that the occurrence of a PSC is favored by the typical geometry in most of these experiments,[3,4] in which the tip is superconducting and the flat is normal.

The analysis of Gesckenbein and Sokol[8] (GS) based on the time-dependent Ginzburg-Landau (TDGL) theory demonstrates that the experimental results may be understood as second-order Josephson effects.[9] These authors have studied the solutions of the TDGL equations under current flow conditions, in the cases of a superconductor separated from a normal metal by an insulating layer (SIN) and superconductor–normal-metal contacts (SN). The results that are applicable to the point-contact experiments are those for the SIN geometry, since coupling between the electrodes must be assumed to be weak.[8,9] In the case of a barrier of low transparency, the superconducting order parameter may be considered unaffected by the contact, and the TDGL equations have only time-independent solutions, which are simple to calculate. The resulting superconducting order parameter in the normal metal decays more rapidly in space as voltage increases. In the normal metal near the junction, there are superconducting and normal components of the current. The superconducting component, which is zero at zero bias, increases very fast with voltage, reaches a maximum apparent critical current, and then decreases more slowly, yielding a nonlinear current-voltage characteristic that resembles that of a standard Josephson effect.

In the experimental works mentioned above,[2–4] point-contact devices typically use a micrometer screw to approach tip to flat. The development of the scanning tunneling microscope (STM) opens the possibility of studying the continuous variation of the barrier strength in a nanoscopic junction as well as the spatial position of the probed area. Several authors have used this tool to study the superconducting gap as a function of spatial position or with high tunneling resistances far from contact.[10] The vortex state in $NbSe_2$ (Ref. 11) has also been studied. Nevertheless, they have concentrated on spatial variations of the superconductivity, but not on a systematic barrier strength variation.

In this paper, we present our results in STM spectroscopy for SN and SS junctions. These experiments have been done using an inertial STM described elsewhere,[12] which allows us get a continuous variation of resistance, with a high degree of control, from about 100 MΩ to 1 Ω. Thus, we can observe the transition between the different regimes described above and get some insight on the intervening processes.

We have used polycrystalline lead, cast from chemically pure lead granules (nominal purity 99.99%) and machined into tips and flats. The tips were Pt-Rh; the Au sample was a single crystal. The surface of the lead sample is scratched clean just before introduction in the microscope. It is exposed to air no more than 15 min. Spectroscopic measurements were carried out ramping the tunneling voltage at a fixed position (feedback interrupted) in 10 ms and the signal was digitized into 1024 points. Current was detected using a variable gain preamplifier (Keithley 428). Conductance curves were obtained by numerical derivation. We concentrated on measuring characteristic curves at different positions and on a continuous variation of the barrier, both advancing and receding the tip. Correlation of topographic and spectroscopic data was not attempted because spectroscopic results were equivalent at all sites probed.

Since the control parameter in our STM unit is the resistance of the junction, we will discuss the obtained results as a function of this parameter. Note that it might seem desirable to use tip-to-flat distance as a parameter, but this is not really known, and besides in the contact regime what is relevant is the variation of contact surface and this reflects directly on the junction resistance.

In the first place, we will consider the system composed of a Pb flat and a Pt-Rh tip. In Fig. 1, we can see the typical evolution of the differential conductance curves as tip-to-flat resistance is varied. Notice that we are plotting the logarithm of the conductance, and consequently we can cover several orders of magnitude in the same graph. For high values of the junction resistance, the conductance is characterized by a dip at zero bias which turns into a hump for resistances of the order of the quantum unit of resistance, $R_q = \hbar/2e^2$=12900 Ω.[13] For resistances of several ohms a sharp zero-bias conductance peak appears. This peak becomes sharper as the resistance diminishes further. It must be remarked that this behavior is not particular to one spot, we have been able to reproduce it in many different sites of the sample, not only qualitatively but quantitatively, and both advancing and retreating the tip. This experiment has been repeated for different Pb flats and Pt-Rh tips, the results being essentially identical. In this figure two regions of the resistance may be distinguished, the first corresponds to values from infinity to the appearance of the zero-bias hump, and the second to the region in which this hump dominates and the sharp zero-bias peak appears.

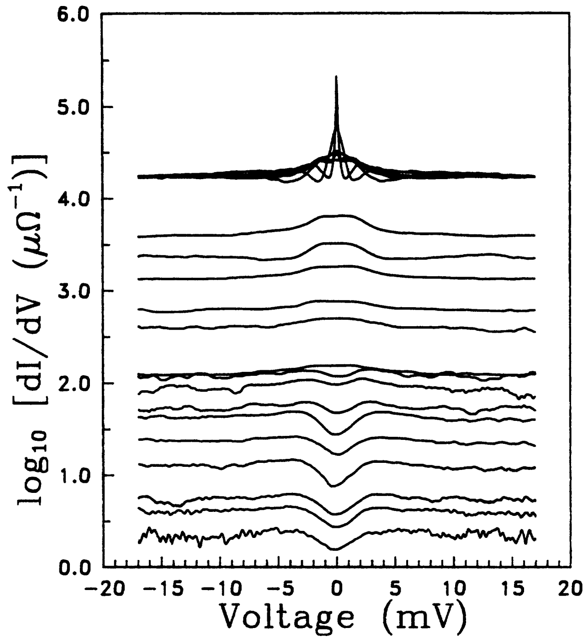

FIG. 1. Experimental conductance curves for Pb (flat) Pt-Rh (tip). Note log vertical scale. Bottom corresponds to high resistance and top to low resistance. Quantum resistance is $\hbar/2e^2$ = 12900 Ω (1.89 in the vertical scale).

The first region can be explained within the BTK theory,[1] as already mentioned. Our experimental curves show strong smearing with respect to those given by the BTK model. We have used a pair-breaking model which phenomenologically incorporates the effects of inelastic scattering substituting E by $E + i\Gamma$ in the Bogoliubov equations in the BTK model. In Fig. 2, we can see the results of this modified BTK model, using Δ=1.3 meV and Γ=0.7 meV, for all the curves. The curve at the bottom is the BCS limit. It can also be obtained using the conventional BCS model with a density of states

$$N(E,\Gamma) = \mathrm{Re}\left\{\frac{E - i\Gamma}{[(E - i\Gamma)^2 - \Delta^2]^{1/2}}\right\}. \qquad (1)$$

It must be remarked that curves in Fig. 2 have only an arbitrary additive constant, which represents the area of the contact.[14]

The value Γ = 0.7 meV fits correctly the conductance at zero bias in the entire range. Both the gap and the Andreev peak are somewhat wider than predicted by the model. Although the reason for such large values of Γ is not clear, this seems to be the case for many (but not for all) of the STM studies of superconductors.[10,11]

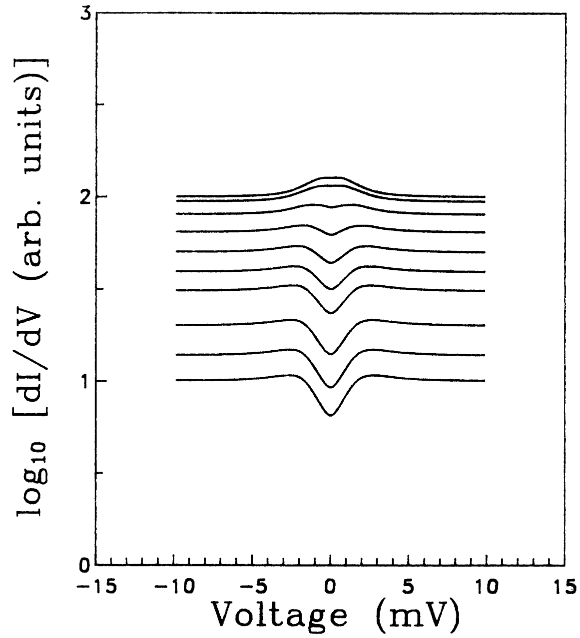

FIG. 2. Model of transition from tunneling to contact using modified BTK model. Note that the, in principle, unknown area would imply an additive constant in the logarithmic plot (but spacing is not arbitrary).

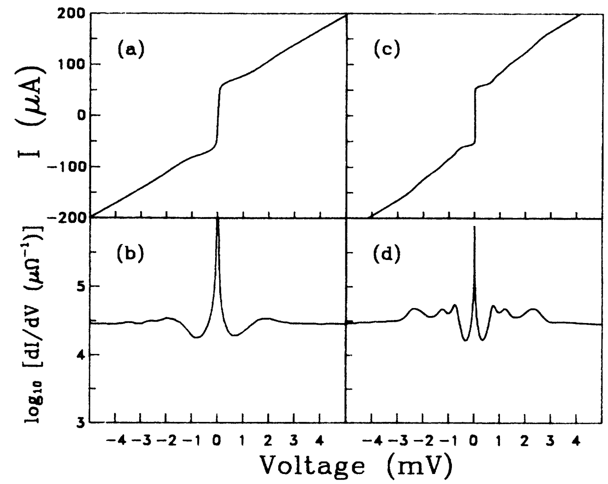

FIG. 3. Experimental curves showing SN Josephson effect and SS Josephson effect. (a) SN *I-V* curve; (b) SN differential conductance (log scale); (c) SS *I-V* curve; (d) SS differential conductance (log scale). Note the subharmonic structure in SS, which is absent in SN.

Pair breaking caused by the large current densities in STM is one of the often invoked reasons for this nonideal behavior, however, we have obtained the same value of Γ for a three orders of magnitude variation of the current. We feel that surface contamination possibly in the form of lead oxide may be the cause.[2,11]

The second region starts with a parallel displacement of the conductance curve that implies an increase in conductance clearly due to the increase in the contact area. When the resistance is about 100 Ω,[15] another contribution to the zero-bias conductance becomes evident in the form of a sharp peak. As mentioned above, two conflicting interpretations of this sharp peak can be found in the literature: one of them considers that the zero-bias peak is due to a second-order proximity-induced Josephson effect (PJE),[9] while the other requires the existence of phase-slip center (PSC) near the junction that would yield a first-order Josephson effect.[7] Discerning between both effects is not easy, because the corresponding characteristic curves are very similar and the real geometry of the nanoscopic junction is not certain.

In order to clarify the origin of this sharp peak we have used several configurations: N-tip(Pt-Rh) vs S-flat(Pb); S-tip(Pb) vs N-flat(Au); and S-tip(Pb) vs S-flat(Pb). These results may be summarized as follows. Pb vs Au yields identical results to Pt-Rh vs Pb (Fig. 1). This speaks against the existence of a PSC, which would be plausible in an S-tip but not in *all* probed areas in an S-flat.[7] Furthermore, a superconducting particle stuck to the normal electrode and giving rise to an effective SS junction is plausible if the tip is normal (due to contact between tip and flat), but not if the tip is superconducting because we can change position of the tip horizontally by about 1000 nm without touching the flat and it is very unlikely that the whole N-flat surface was covered by a superconducting particle from the tip. We have repeated the experiment at different locations approaching the sample from macroscopical distances, which implies probing different zones of the sample. We could also vary tip-to-flat distance gradually and find that curves like those in Fig. 1 can be taken in a completely reproducible manner either approaching or receding from the sample. Large resistance curves obtained after touching the sample are identical to those obtained when carefully avoiding contact. Clearly, the tip is either always clean or always contaminated by the sample.

Furthermore, our results for Pb vs Pb show a different behavior at the SN junctions. For microcontacts of small resistance, subharmonic effects can be clearly seen (see

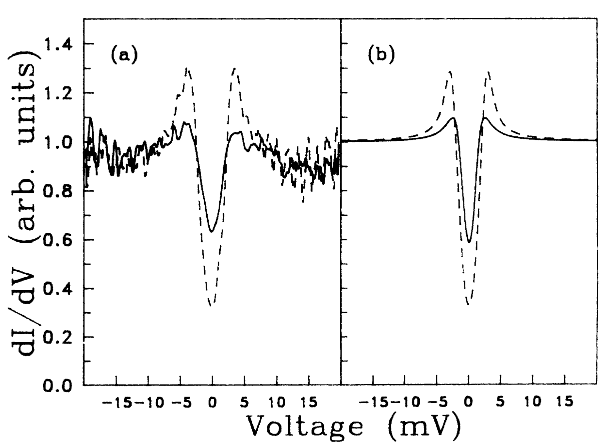

FIG. 4. SN gap compared to SS gap, experimental and theoretical. (a) Experimental differential conductance curves: SS (dotted line), SN (solid line); (b) theoretical differential conductance curves: SS (dotted line), SN (solid line).

Fig. 3). The onset of this SS supercurrent is for about 800 Ω.[16] For large resistances the resulting conductance curves can be fitted using BCS theory for SS tunneling. It must be remarked that the same value of the pair-breaking parameter, $\Gamma = 0.7$ meV (see Fig. 4) is used to fit the experimental curves.

In summary, the ability of STM to change tip to flat distance (or pressure when they are in contact) gradually, makes possible the clear distinction of a SS junction from a SN junction, both in the tunneling and in the point-contact regime. Consequently, we believe that our experiments show in a convincing manner that in junctions in which one of the electrodes is normal and the other superconducting, the observed zero-bias peak at low resistance is not due to the existence of a phase-slip center in the superconducting electrode or near the NS interface, and is consistent with the interpretation given by Gesckenbein and Sokol[8] and Han, Cohen, and Wolf[9] as a second-order proximity-induced Josephson effect.

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[14] This area may be estimated as follows. When the barrier collapses the Andreev peak reaches its maximum height (about 7 kΩ in Fig. 1). Contact is then metallic and we may use the Sharvin expression for the resistance, $R_S = \rho l/4a^2$, where ρ is the resistivity of the bulk, l is the mean free path, and a is the radius of the contact [see, for example, A.M. Duif, A.G.M. Jansen, and P. Wyder, J. Phys.: Condens. Matter **1**, 3157 (1989)], to estimate the area, which results in $a = 0.26$ nm (or the area occupied by two atoms). This estimate may not be valid for such a small size but the resistance for a one-atom contact should be of the order of the quantum resistance unit $R_q = 12900$ Ω (Ref. 13).

[15] This corresponds to $a = 2$ nm or about 120 atoms.

[16] This corresponds to $a = 0.8$ nm or 15 atoms.