Evidence for Suppression of Superconductivity by Spin Imbalance in Co-Al-Co Single-Electron Transistors

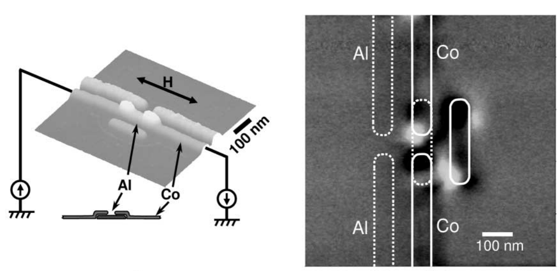
C. D. Chen,[1,*] Watson Kuo,[1,2] D. S. Chung,[1] J. H. Shyu,[1,3] and C. S. Wu[1]
[1]*Institute of Physics, Academia Sinica, Nankang 115, Taipei, Taiwan, Republic of China*
[2]*Department of Physics, National Tsing-Hua, Hsin-Chu 300, Taiwan, Republic of China*
[3]*Department of Physics, National Taiwan University, Taipei, Taiwan, Republic of China*
(Received 31 March 2001; published 14 January 2002)

Spin imbalance can lead to suppression of superconductivity. We report the phenomena manifesting this effect under spin-polarized quasiparticle currents in ferromagnet-superconductor-ferromagnet single-electron transistors. The measured superconducting gap as a function of magnetic field reveals a dramatic decrease when the magnetizations of the two leads are misaligned. The effect of suppression increases with increasing source-drain voltage. A comparison with theoretical calculations is presented. This method may render it applicable to control superconductivity at low temperatures within low fields.

DOI: 10.1103/PhysRevLett.88.047004 PACS numbers: 74.25.Ha, 75.75.+a, 85.35.Gv, 85.75.–d

The interplay between superconductivity and magnetism has been a topic of interest for many years. Of particular interest are recent experiments on the ferromagnet/superconductor (FM/SC) junctions with high T_C superconductors [1] in which a decrease of the supercurrent by nonequilibrium spin density was demonstrated. Furthermore, theoretical studies [2,3] also suggest that the spin imbalance in a superconductor can lead to suppression of superconductivity. In a double tunnel junction containing a normal metal or superconductor sandwiched between two ferromagnets, both injection of polarized current and spin imbalance are possible, thus providing an ideal testing ground for this theory. When the magnetic moments of the two ferromagnetic leads are in opposite orientations, electrons in the source electrode with majority spins experience a normal tunnel resistance to enter the central electrode, but encounter an enlarged resistance to tunnel out to the drain electrode. Consequently, the minority spins in the central electrode would tunnel out more easily than the majority spins, resulting in a spin imbalance. This induces a chemical potential difference in the central electrode, which, in turn, would decrease the tunneling out rate of the minority spins. In the equilibrium state, this potential difference is $\delta\mu = PeV/2$ for a normal metal central electrode, where P is the polarization of the ferromagnetic leads, and V is the voltage across the sample. For a superconducting central electrode, this difference gives rise to pair breaking, and suppresses superconductivity, in the same way as the Zeeman effect does to superconductivity in the paramagnetic limit.

In this paper, we report direct observation of the superconducting gap suppression using Co/Al/Co (FM/SC/FM) double tunnel junctions. This effect can be turned on and off by manipulating mutual orientations of magnetic moment of the two Co leads. Figure 1a shows an atomic force microscope (AFM) picture of a measured sample and its biasing circuit. A gate electrode (not shown in the image) located about 2 μm away is used to tune the potential of the central electrode. The samples were fabricated by standard electron-beam lithography techniques and by the two-angle evaporation method [4]. A thin native Al_2O_3 layer between the Al island and the Co electrodes acts as a tunnel barrier. This native oxide layer was formed by introducing 50 mbar of pure oxygen gas into the chamber, after deposition of Al and prior to deposition of Co, for about 2 min, with the sample at room temperature. Aluminum is a good candidate to serve as the superconductor for this purpose, not just for its long spin lifetime [5] allowing the full range of spin effects to be studied, but also for its high quality native Al_2O_3 barrier which was shown to have no spin-flip tunneling processes [6]. Our electron gun deposited Al islands have a superconducting transition temperature T_C of about 2 K. The samples were measured using a dilution refrigerator, and the magnetic field was applied along the Co leads, i.e., parallel to the long edge of the Al island. Because of the small thickness (≈25 nm) of the Al island and of the alignment with the magnetic field, the critical magnetic field H_C for the Al islands is about 21 kOe.



(a) (b)

FIG. 1. (a) AFM and (b) magnetic force microscope (MFM) images of a sample. Because of a two-angle evaporation technique used for fabrication of the samples, there are redundant, electrically unconnected structures aside the measured device. The inset in (a) illustrates the cross section of the island and junctions. The magnetic image was obtained using MFM with low magnetic stray field and high coercivity CoPt tips. The Co and Al electrodes are indicated with solid and dashed curves, respectively.

Because of the small size of the tunnel junction, the charging energy E_C associated with it is large [7]. The current-voltage characteristics measured at low temperatures display, in addition to the superconducting gap $2\Delta_o/e$, a pronounced Coulomb gap $2E_C/e$. The *IV* characteristics measured at $T \sim 40$ mK for $V_g = e/2C_g$, where the Coulomb gap is suppressed to a minimum, are shown in the inset of Fig. 2 with the Al island being in the superconducting state ($H = 0$, labeled with "I") and in the normal state ($H = 25$ kOe, labeled with "II"). Here, V_g is the gate voltage and C_g is the capacitance between the gate electrode and the central island. From a plot of zero-field *IV* characteristics as a function of gate voltages (i.e., the "Coulomb blockade parallelogram" [7]), we estimate a superconducting gap Δ_o of approximately 250 μeV, and a charging energy E_C of $\sim$100–150 μeV.

Throughout this study, samples were symmetrically current biased with respect to the ground. We applied a parallel magnetic field to decrease the size of the superconducting gap, and monitored this decrease by measuring the voltages (see inset of Fig. 2). The temperature during the measurement was about 250 mK. As shown in Fig. 2, when the field ramps down from +25 to −25 kOe, we find a sharp voltage drop at a field of around −1.5 kOe; when the field is reversed and ramped up from −25 to +25 kOe, the voltage drops again at a field around +1.5 kOe, yielding a symmetrical $V(H)$ pattern with respect to zero field. This hysteresis is reproducible, and similar results are found for other samples. The hysteresis behavior is very similar to the hysteresis seen in the tunneling magnetoresistance of a Co/carbon nanotube/Co system [8]. By a token of this similarity, it is reasonable to argue that at small fields ($H = \pm 1.5$ kOe for the sample of Fig. 1) the magnetization of the two leads are misaligned, whereas, at a field of $H = 25$ kOe, the magnetization saturates and aligns parallel to the applied fields. If the two leads were made of nonmagnetic materials, it would be an arch-shaped $V(H)$ curve with no voltage dips, and the height of the arch would be approximately $2\Delta_0/e$. These dips in $V(H)$ curves indicate a reduction of the superconducting gap of the Al island.

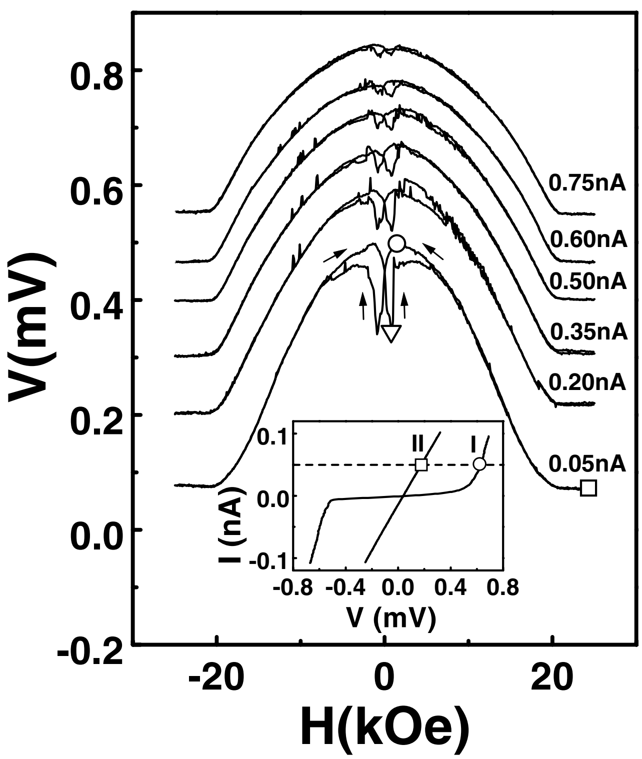


FIG. 2. The inset shows the measured *IV* characteristics at $V_g = e/2C_g$ and $H = 0$ (labeled with "I"), and $H = 25$ kOe (labeled with "II"). The horizontal dashed line indicates the constant-current load line for $V(H)$ measurements. The main panel shows measured $V(H)$ curves for several selected bias currents at $T \approx 250$ mK. The small arrows along the 50 pA curve indicate the ramping direction of the applied field. For each curve (shown, for example, for $I = 50$ pA curve), the voltage values at the top of the arch (marked with ○), at $H = 25$ kOe (marked with □), and at the dip (marked with ▽) are used to reconstruct *IV* characteristics shown in Fig. 3, labeled with I, II, and III, respectively. The points in the $V(H)$ curve marked with ○ and □ are also indicated in the inset.

In Ref. [8], the formation of the misalignment state was attributed to local fluctuations of magnetization orientation. Figure 1b shows the domain structures of the Co electrodes imaged by magnetic force microscopy. Even the exact knowledge of the magnetic structure is not known; it is likely that the ends of the Co electrodes (lying on top of the Al island) form single domains, because they are topographically higher than the rest of the electrodes, and their size is small. We therefore interpret the measured hysteresis as simply a sign of magnetization reversal of a single domain.

The theory by Takahashi, Imamura, and Maekawa [2] predicts a decrease of Δ_A with increasing source-drain voltage V, with Δ_A being the superconducting gap when the two ferromagnetic leads are in antiferromagnetic alignment. In this alignment, excess spins accumulate in the superconducting island, and the chemical potential of the majority and minority spins are shifted oppositely by $\delta\mu$ from the equilibrium state. This chemical potential difference plays the role of pair breaking energy, leading to a suppression of superconducting gap from Δ_o to Δ_A. In the equilibrium state, $\delta\mu$ increases with V, and, consequently, Δ_A decreases with V. Theoretically, this dependence of Δ_A on V is determined by combining $\Delta_A(\delta\mu)$ and $\delta\mu(eV)$ functions, and self-consistently calculating the values of $\delta\mu$, eV, and Δ_A [2]. In the experiments, however, because Δ_A varies with source-drain voltage, it cannot be derived by simply identifying the peak position of dI/dV vs the V plot [9]. As we describe below, along with the help of a simulation for *IV* characteristics for various superconducting gaps, the Δ_A can be unambiguously determined.

Figure 2 also shows $V(H)$ curves taken at various bias currents. One can use voltage data at the top of the arch (fairly close to $H = 0$, marked with ○), at $H = 25$ kOe (marked with □), and at the dip ($H \approx 1.5$ kOe, marked with ▽) of each curve to reconstruct three *IV* characteristics as shown in Fig. 3, labeled, respectively, with I, II, and III. Curves I and II, respectively, are for the island in the superconducting state and in the normal state. Curve III is for the case of a superconducting island, with the two leads' magnetizations being misaligned. Curve I can be fitted by simulation of a normal metal/superconductor/normal metal single-electron transistor. The simulation is based on the master equation [10] in equilibrium state, with an assumption that only one electron tunnels at a time. From this fitting, one obtains a good estimate of sample parameters, including the capacitances and the resistances of the two tunnel junctions and, most importantly, the zero-field superconducting gap Δ_o of the Al island. The fitted Δ_o value is approximately 260 μeV, agreeing well with the gap estimate from the plot of the Coulomb blockade parallelogram. The parameters for this particular sample are as follows: $R_{\text{source}} = R_{\text{drain}} = 237$ kΩ, $C_{\text{source}} = 360$ aF, $C_{\text{drain}} = 300$ aF, $C_g = 0.47$ aF, and $V_g = 160$ mV. A temperature of 250 mK is used for this fitting. The obtained junction parameters from the fittings of curve I can then be used to generate an *IV* curve to compare with curve II, that is, for an all-normal-metal (N/N/N) transistor. A small deviation may be due to tunneling processes that are not included in the simulation. For example, it is known that, for $V_g = e/2C_g$, there is a knee at $V = 2E_C/e = 0.24\boldsymbol{eV}$, below which the sequential tunneling process suppresses the differential conductance by a factor of 2. However, in reality, this feature may be smeared probably due to Zener tunneling to the upper band [11].

Now we should proceed to discuss the voltage dependence of Δ_A. In the antiferromagnetic alignment, the effective drain tunneling resistance R_{DA} increases by a factor of $1 + P^2/1 - P^2$. For the case of the Co electrodes, the reported P value is about 0.4 [9]. Taking into account this correction, along with the sample parameters obtained above, one can calculate a series of *IV* characteristics for various superconducting gaps Δ_A. In Fig. 3, we plot the calculated curves together with curves I–III. Each calculated curve for an assigned Δ_A crosses curve III at a point, and, hence, produces a set of (Δ_A, V) data. Plotting the data set for all crossing points, we obtain the dependence of Δ_A on source-drain voltage V, depicted in Fig. 4. In the same figure, we also show this dependence obtained for another sample with similar parameters. In accordance with the theory of Ref. [2], in which the charging effect is not considered, we calculate $\Delta_A/\Delta_o(V)$ curves for $P = 0.4$, and $T = 250$ mK ($= 0.125T_C$), and the result is shown (as a dashed curve) in Fig. 4. The charging effect is known to lead to two distinct consequences, namely, a sequential tunneling process at low voltages and a constant offset voltage E_C/e at high voltages. By incorporating the master equation for sequential tunneling, we also calculate the $\Delta_A/\Delta_o(V)$ curves using our sample parameters, for, again, $P = 0.4$ and $T = 0.125T_C$, and the result is shown as a solid curve in Fig. 4. At high voltages, the offset in the *IV* characteristic leads to a shift in the Δ_A/Δ_o dependence on V at $V = E_C/e$, which for both measured samples is about 120 μV ($\approx 0.48\Delta_0/e$). Details of the calculations will be published elsewhere.

Our analysis for both samples shows qualitatively the same type of $\Delta_A(V)$ dependence: Δ_A increases with V

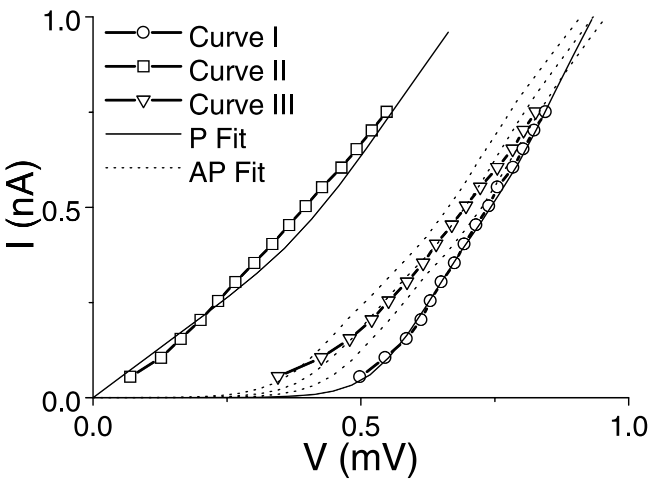


FIG. 3. Bold curves: The *IV* characteristics as reconstructed from Fig. 2. The two solid curves are calculated, respectively, for the superconducting and normal states, and the dashed curves are calculated for various Δ_A values (shown in the figure are 220, 200, 180 μeV), as described in the text. The voltage values of the crossing points between curve III and the calculated curves as a function of Δ_A are plotted in Fig. 4.

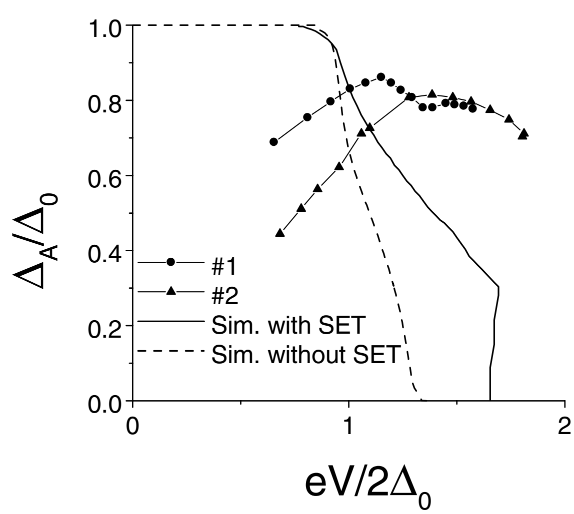


FIG. 4. Δ_A/Δ_o as a function of voltage. Curves with symbols are data from two measured samples. Calculated curves for $T = 250$ mK with (solid curve) and without (dashed curve) charging effect are also plotted for comparison.

at small voltages, and decreases at larger voltages. Ideally, at low temperatures and low voltages, absence of quasiparticle tunneling should lead to a Δ_A/Δ_o value close to 1. Various origins can produce excess quasiparticle current, which are not taken into account in our simulation. This excess current, in our analysis, may render lower Δ_A values than expected. One possible source for such excess current is the cotunneling process [7]. In this process, excess current increases at voltages close to Δ_A/e, and strongly influences IV curves with smaller Δ_A values; hence, it is responsible for the decrease of Δ_A/Δ_o values at lower voltages. On the other hand, at high voltages, Δ_A/Δ_o values for both samples are larger than the calculated solid curve, i.e., the suppression is less effective. The discrepancy probably arises from an overly long energy relaxation time, and a short spin relaxation time in our small Al island, which will diminish the effect of spin imbalance. The imbalance of spins is a nonequilibrium process, but, under present theoretical calculations, we assume a sufficiently long spin relaxation time and a short energy relaxation time. A necessary condition for the imbalance is that the spin relaxation time τ_s is longer than the time τ_t between two successive tunneling events. A rough estimate of τ_t is e/I [12], which in our experiments is $\sim$0.1–1 ns. In our Al islands, τ_t may be somewhat shorter than the reported spin relaxation time τ_s of about 10 ns in a bulk Al [5,13]. This spin relaxation process should also be responsible for a smeared gap suppression.

Despite the clear deviations between the calculated and the derived $\Delta_A(V)$ dependence, and possible errors that may arise from uncertainties involved in deriving the $\Delta_A(V)$ dependence, this experiment suggests a suppression of Δ_A with increasing bias voltage. This is not the case of the magnetoquenched superconducting valves [14], where the superconductivity is suppressed by the stray field emitted from the two antiferromagnetically aligned leads, and is bias voltage independent. In our case, the stray field is presumably small due to the smallness of the single domains at the ends of electrodes, and the influence on the superconductivity of the Al island should be small compared with the effect of spin imbalance. Nevertheless, further study to separate these two effects would be beneficial.

In conclusion, the superconducting gap of a small Al island, when incorporated in a single electron transistor structure with two Co electrodes, was found to be strongly suppressed by polarized quasiparticle injection from the ferromagnetic leads if the magnetizations of the two leads are misaligned. The effect appears to be caused by pair breaking associated with spin imbalance in the superconducting island. This experiment provides a new method of controlling superconductivity.

Fruitful discussions with Y. D. Yao, S. K. Yip, G. Y. Guo, and S. F. Lee are gratefully acknowledged. We also thank S. H. Liou of University of Nebraska for his help in magnetic domain images and for providing stimulative discussions. This research was partly funded by the National Science Council No. 89-2112-M-001-033.

*Email address: chiidong@phys.sinica.edu.tw

[1] N.-C. Yeh *et al.,* Phys. Rev. B **60**, 10 522 (1999); Kiejin Lee *et al.,* Appl. Phys. Lett. **75**, 1149 (1999); V. A. Vasko *et al.,* Phys. Rev. Lett. **78**, 1134 (1997); Z. W. Dong *et al.,* Appl. Phys. Lett. **71**, 1718 (1997).

[2] S. Takahashi, H. Imamura, and S. Maekawa, Phys. Rev. Lett. **82**, 3911 (1999); S. Takahashi, H. Imamura, and S. Maekawa, J. Appl. Phys. **87**, 5227 (2000).

[3] Zhiming Zheng, D. Y. Xing, Guoya Sun, and Jinming Dong, Phys. Rev. B **62**, 14 326 (2000).

[4] G. J. Dolan, Appl. Phys. Lett. **31**, 337 (1977).

[5] Mark Johnson and R. H. Silsbee, Phys. Rev. B **37**, 5326 (1988).

[6] R. Meservey, P. M. Tedrow, and R. C. Bruno, Phys. Rev. B **11**, 4224 (1975).

[7] For charging effects in general, see, for example, *"Single Charge Tunneling,"* edited by H. Grabert and M. H. Devoret, NATO ASI, Vol. 294 (Plenum, New York, 1992). For magnetic single electron transistors, see H. Imamura, S. Takahashi, and S. Maekawa, Phys. Rev. B **59**, 6017 (1999); S. Takahashi and S. Maekawa, Phys. Rev. Lett. **80**, 1758 (1998); A. Brataas, Yu. V. Nazarov, J. Inoue, and Gerrit E. W. Bauer, Phys. Rev. B **59**, 93 (1999); J. Barnas and A. Fert, Phys. Rev. Lett. **80**, 1058 (1998).

[8] K. Tsukagoshi, B. W. Alphenaar, and H. Ago, Nature (London) **401**, 572 (1999).

[9] R. J. Soulen *et al.,* J. Appl. Phys. **85**, 4589 (1999); R. Meservey and P. M. Tedrow, Phys. Rep. **238**, 173 (1994).

[10] For the computer program algorithm, see H. Dalsgaard Jensen and John M. Martinis, Phys. Rev. B **46**, 13 407 (1992). For a formula of the tunneling rate in a normal metal/superconductor junction, see I. Giaever and K. Megerle, Phys. Rev. **122**, 1101 (1961).

[11] D. V. Averin and K. K. Likharev, in *Mesoscopic Phenomena in Solids,* edited by B. L. Altshuler, P. A. Lee, and R. A. Webb (North-Holland, Amsterdam, 1991), p. 192.

[12] From the continuity equations for electric charges and spins, one can see that in the steady state the spin imbalance $(N_\uparrow - N_\downarrow)$ is related to the tunneling rates for, say, up spins $\Gamma_{l\uparrow}$ and $\Gamma_{r\uparrow}$ on the left and right junctions, respectively, as $(N_\uparrow - N_\downarrow)/\tau_s = \Gamma_{l\uparrow}(\delta\mu + V/2) - \Gamma_{r\uparrow}(\delta\mu - V/2)$. In our experiments, the transistors were operated in the sequential tunneling regime (only one electron can tunnel at a time), and, hence, the tunneling rate is on the order of e/I, which is to be compared with $1/\tau_s$.

[13] In making this comparison, we also assumed that τ_s for quasiparticles in the superconducting state is the same as that in the normal state.

[14] T. W. Clinton and Mark Johnson, Appl. Phys. Lett. **70**, 1170 (1997); J. Appl. Phys. **85**, 1637 (1999).