Paramagnetic reentrant effect in high purity mesoscopic AgNb proximity structures

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(February 2000)

We discuss the magnetic response of clean Ag coated Nb proximity cylinders in the temperature range $150 \mu K < T < 9$ K. In the mesoscopic temperature regime, the normal metal-superconductor system shows the yet unexplained paramagnetic reentrant effect, discovered some years ago [P. Visani, A. C. Mota, and A. Pollini, Phys. Rev. Lett. 65, 1514 (1990)], superimposing on full Meissner screening. The logarithmic slope of the reentrant paramagnetic susceptibility $\chi_{\text{para}}(T) \propto \exp\left(-L/\xi_N\right)$ is limited by the condition $\xi_N = nL$, with $\xi_N = h\nu_F/2\pi k_B T$, the thermal coherence length and $n = 1, 2, 4$. At the lowest temperatures, $\chi_{\text{para}}$ compensates the diamagnetic susceptibility of the whole AgNb structure.

PACS numbers: 74.50.+r, 74.80.-g

In recent years, there has been extensive experimental and theoretical work in the field of mesoscopic systems, including superconducting structures in proximity with normal metals [1]. In particular, the paramagnetic reentrant phenomenon [2] has received a wide interest, mainly because of being promoted by the recent understanding of the high-temperature diamagnetic response of rather clean normal-metal-superconductor (NS) proximity structures [3] in the context of the quasiclassical Eilenberger theory including elastic scattering [4]. In this Letter, we discuss the very low temperature reentrant behavior of two of the AgNb samples of Ref. [5], covering a larger mesoscopic regime with respect to previous measurements [6].

Recently, two Letters [5,6] have addressed the origin of paramagnetic currents in NS systems, which might lead to an understanding of the paramagnetic reentrant phenomenon. The work of Bruder and Imry [5] is based on the presence of non-Andreev-reflecting semiclassical trajectories at the outer surface of a nonsingly connected proximity system (glancing states), which carry predominantly paramagnetic currents. This work has been subject to debate because of the small magnitude [5]. A different, more elaborate approach by Fauchère et al. [6] assumes a net repulsive interaction in the noble metals. The $\pi$ shift of the order parameter at the NS interface then leads to a paramagnetic instability of Andreev pairs.

The first work reflects the cylindrical geometry of our NS system, but it does not address the experimental signatures of the paramagnetic reentrant, namely absolute value of order 1, temperature dependence, nonlinearity, hysteresis, or dissipation [4]. The latter three features might give evidence for a spontaneous magnetization in the samples, as proposed in Ref. [5]. However, the second theoretical approach [6] does not reach beyond qualitative accordance with our experiment.

Here we discuss an investigation of the paramagnetic reentrant effect extending to the $\mu$K region. By covering five decades in temperature, we have been able to extract the correct temperature dependence of the NS proximity structure below $T_c$. Over a large mesoscopic regime, the cylindrical structure clearly displays different levels of coherence along integer multiples of the wire perimeter $L$. Unfortunately, neither of the two present theories [5,6], which discuss different sources of paramagnetism in NS structures, obtain the correct absolute value as well as the $T$ dependence of the paramagnetic reentrant effect.

The two samples reported here are ensembles of cylindrical wires with a superconducting core of soft niobium ($R_{\text{RRR}} = 300$) concentrically embedded in a normal-metal matrix of 6N silver. Their total diameter was mechanically reduced by several steps of swagging and co-drawing [8] to final values 41 $\mu$m and 23 $\mu$m, with normal layer thicknesses $d_N = 5.5 \mu$m and 3.3 $\mu$m (the ratio $d_N/L$ is approximately the same), respectively. The samples were

![Graph](attachment:graph.png)

FIG. 1. Magnetic susceptibility $\chi_N(T)$, between 150 $\mu$K and 9 K. For both samples we show $\chi_{ac}(T)$ (+) and $\chi_{dc}(T)$ (●). The arrows mark the direction of $T$-changes.
annealed after the last drawing, and values of the mean free path $\ell_N \sim (0.5$–$0.8)d_N$ were obtained. For more details see Refs. 3,8.

Extensions of the measurements to $\mu$K temperatures were performed at the ultralow temperature (ULT) facility at the University of Bayreuth. There, an experimental setup was installed for inductive measurements using an rf–SQUID sensor. Magnetic fields were applied along the axis of the wires.

For the ULT experiments, we took parts of the wire bundle measured in our dilution refrigerator 2,3, and glued them with GE 7031 varnish to high purity gold foils tightly attached to a silver finger, in good electrical contact with the Cu demagnetization stage 10. Thus, about 200 wires were mounted. Temperatures were measured with a Pt pulsed NMR thermometer 14.

In the following we report on the temperature dependent magnetic susceptibility of our relatively clean silver-niobium samples 3AgNb $[d_N = 5.5 \mu m]$ and 5AgNb $[d_N = 3.3 \mu m]$. In Fig. 1 the total magnetic susceptibility is shown as a function of temperature. We show $\chi_{ac}(T)$ between 4 mK and 9 K, measured in our dilution refrigerator with field amplitude $H_{ac} = 33$ mOe and frequency $\nu = 80$ Hz, as well as $\chi_{dc}(T)$ at constant $H_{dc}$ at ULT and LT.

At temperatures below the critical temperature of Nb $T_c = 9.2$ K, the magnetic susceptibility of the N layer exhibits diamagnetism induced through Andreev reflection at the highly transparent NS interface. At lower temperatures, it develops almost total Meissner screening in the Ag layer 1. Below $T_r \sim 100$ mK the signature of reentrance is observed both in $\chi_{ac}(T)$ 2 and $\chi_{dc}(T)$, with the development of an additional paramagnetic susceptibility $\chi_{para}(T)$, such that $\chi_N(T) = \chi_{dia}(T) + \chi_{para}(T)$.

For sample 3AgNb, the susceptibility $\chi_N$ saturates below $T_{sat} \approx 400 \mu K$, displaying a complete cancellation of the induced diamagnetic susceptibility in N, such that only the diamagnetism in S seems to remain. For sample 5AgNb the susceptibility $\chi_N$ shows saturation below $T_{sat} \approx 800 \mu K$ at a paramagnetic value $4\pi \chi_{N}^{M} \approx 1$, indicating a complete cancellation of the total diamagnetic susceptibility in N plus S.

The anomalously large magnitude of the paramagnetic reentrant susceptibility $\chi_{para}$ at ultralow temperatures is rather intriguing. This magnitude, and particularly its dependence on the sample size, is not explained by existing theories. At this moment, it is not clear, if paramagnetic reentrance is (i) an intrinsic effect of mesoscopic NS proximity structures in the very low temperature limit or (ii) the result of two independent phenomena. In the latter case the selection of the NS materials could be important.

A detailed inspection $\chi_N$ around its minimum reveals that, the reentrance temperature $T_r$ is decreased under a field $H_{dc} = 0.2$ Oe. The ac and dc curves show reentrant temperatures $T_r = 97$ mK and $T_r^H = 83$ mK for sample 3AgNb ($T_r = 149$ mK and $T_r^H = 113$ mK for sample 5AgNb), respectively.

Neglecting the weak-field effect on $T_r$, $\chi_{dc}(T)$ matches $\chi_{ac}(T)$ for sample 3AgNb (5AgNb) between 15 mK (30 mK) and 1 K. This is rather noticeable, considering the strong nonlinearity in magnetic field discussed below.

Above $T \approx 1$ K, $\chi_{dc}(T)$ deviates from $\chi_{ac}(T, H_{ac} \approx 0)$, due to the depression of the weak induced Andreev pair potential by finite fields. Below 15 mK (30 mK), $\chi_{dc}(T)$ agrees with $\chi_{ac}(T)$ only on measurements done under cooling. Below 15 mK (30 mK) the warming up curve of $\chi_{dc}(T)$ lies above the cooling curves, corresponding to stronger paramagnetism adding to the full Meissner screening.

In our arrangement, samples are in rather good thermal contact with the Cu demagnetization stage and the Pt NMR thermometer. Indeed, the measured thermal relaxation times at the lowest temperatures remain below 1000 s. Nevertheless, the susceptibility shows hysteresis. We have found the highest levels of paramagnetic reen-
transition only after allowing the NS system to remain at much below their saturation temperatures for long periods of time (one week or longer).

Fig. 3 shows the reentrant paramagnetic susceptibility \( \chi_{\text{para}}(T) \) below \( T_r \). The data exponentially increases as \( \chi_{\text{para}}(T) = A \exp \left( -T/T^* \right) \). For sample 3AgNb [Fig. 2(a)], the prefactor \( A_1 = 0.24 \) and characteristic temperature \( T^*_1 = 13 \text{ mK} \) were obtained from \( \chi_{\text{ac}}(T) \). The cooling and warming \( \chi_{\text{dc}}(T) \), above \( \approx 13 \text{ mK} \) reproduce well the behavior of \( \chi_{\text{para}} \) observed in \( \chi_{\text{ac}}(T) \). Near \( \approx 13 \text{ mK} \), \( \chi_{\text{dc}} \) displays a kink, leading to an approximate doubling of the logarithmic slope, with \( T^*_2 = 5.5 \text{ mK} \sim T^*_1/2 \) and a prefactor \( A_2 = 1.1 \).

For sample 5AgNb [Fig. 2(b)], the prefactor \( A_1 = 0.4 \) and characteristic temperature \( T^*_1 = 20 \text{ mK} \) were obtained from \( \chi_{\text{ac}}(T) \). Again the cooling and warming \( \chi_{\text{dc}}(T) \) above \( \approx 30 \text{ mK} \) follow the \( T \) behavior of \( \chi_{\text{ac}}(T) \). The curves show a kink in the susceptibility, displaying a doubling of the logarithmic slope, with \( T^*_2 = 11 \text{ mK} \sim T^*_1/2 \) for the second line. In addition, around \( \approx 14 \text{ mK} \), a second doubling of the logarithmic slope of \( \chi_{\text{dc}}(T) \) occurs, with \( T^*_3 = 4.8 \text{ mK} \sim T^*_2/2 \) for the third line.

The coherence length of the Andreev pairs \( \xi_N \) in Ag, obtained from our breakdown field measurements, is in agreement with the clean limit theory, \( \xi_N = \hbar v_F/2\pi k_BT = 1.69 \mu\text{m}/T(K) \). At the temperature of the first kink in \( \chi_{\text{para}} \), \( \xi_N(T) \) reaches approximately the value of a single wire’s circumference \( L = 130 \mu\text{m} \) \( (L = 72 \mu\text{m}) \). In Fig. 2 we have indicated by vertical arrows the temperature at which the equality \( \xi_N(T) = L \) is met. For sample 5AgNb, at the temperature of the second kink it is \( \xi_N(T) = 2L \). The values of \( T^*_2 \), \( T^*_3 \), and \( T^*_3 \), as well as the position of the kinks, which are located approximately at \( T^*_1 \), \( T^*_2 \), give evidence for different levels of quantum coherence on the mesoscopic length scale \( L \). The temperatures \( T^* \), which characterize the different levels as well as the kinks can be written as \( T^* \approx \hbar v_F/2\pi k_B nL \), or in the equivalent form \( \xi_N(T^*) = nL \), with \( n = 1, 2, 4 \). The reentrant paramagnetic susceptibility then is

\[
\chi_{\text{para}} = A_n \exp \left[ -\frac{nL}{\xi_N(T)} \right], \quad \text{with } n = 1, 2, 4. \tag{1}
\]

This characteristic behavior is not obtained by present theories. The correct theory of the paramagnetic reentrant effect should describe the susceptibility in accordance with Eq. (1).

For a theoretical understanding of the origin of the reentrant effect, it is important to investigate the susceptibility under fields, as well as the magnetization. In the following we discuss isothermal ac susceptibility measurements of sample 5AgNb, as a function of magnetic field [Figs. 3 and 4(a)]. The isothermal susceptibility \( \chi_N(H) \) shows nonlinear behavior in the entire field regime. At \( 7 \text{ mK} \) two curves are shown, the first measurement directly after cooldown, and the second one

\[
\sim 10^6 \text{ s} \text{ after the first one. The lower of the two curves at } 7 \text{ mK} \text{ shows pronounced hysteresis of the reentrant part at fields below } 20 \text{ Oe. Near } 20 \text{ Oe the specimen screens the magnetic flux most effectively, before the well known}
\]
magnetic breakdown transition occurs at its field dependence remains to be obtained by the- 

ing. At this sample, the transition is nearly temperature independent for \( T < 50 \text{ mK} \). However, the minimum suscep-
tibility before the breakdown is far from complete screen-
of order 1, the characteristic dependence of the magnetic

ity. Indeed, measurements on a single wire are desirable.

For very small dc magnetic fields, the magnetic sus-
ceptibility grows rather steeply. At higher fields, that
increase slows down, the susceptibility reaching its max-
imum value at about 2.5 Oe, before turning towards less
paramagnetic values. As the temperature is increased,
the curves show a less pronounced maximum at about
2.5 Oe and reduced hysteresis. The detailed behavior at
very low fields differs from the results of similar measure-
ments, performed on a bigger wire bundle, reported in
Ref. [2]. Possibly, the different geometrical arrangement
of the wires in the bundles affect the average susceptibil-
ity. Indeed, measurements on a single wire are desirable.

Cycling the magnetic field in both directions at 7 mK
[Fig. 3(a)], we observe two low-field peaks of \( \chi_N(H) \),
which are displaced symmetrically from zero (residual
field \( < 2 \text{ mOe} \)). However, the curves are not symmet-
ric below 20 Oe, displaying a reduction of hysteresis after
each half-cycle. Furthermore, after cooldown from above
50 mK, cycling the field at 7 mK, and waiting for \( \sim 10^6 \text{s} \),
the low-field peak at about 2.5 Oe grows up. This can be
observed in the upper curves in Figs. 3 and 3(a). After
cycling the field and waiting, the whole system crosses to
a more stable state, with more pronounced paramagnetic
susceptibility.

Hysteresis and nonlinearity are also observed in dc
magnetization curves, e.g. shown for a full cycle at 7 mK
in Fig. 3(b). For the second half-cycle, nonlinearity be-
comes less pronounced, in accordance with our findings
in the susceptibility curves. The measured magnetization
lies between two lines at fields below the breakdown tran-
sition. At low fields one observes clearly the deviation of
the magnetization from the induced Meissner screening.
At higher fields the curve asymptotically approaches the
drawn line indicating linear Meissner-like behavior plus
a constant paramagnetic magnetization. The \( H = 0 \)
intersections of the lines suggest a field-independent mag-
etization \( 4\pi M_0 \approx 1 \text{ G} \). It is interesting to notice that a
spontaneous magnetization at the same order was found
in the theoretical model of Fauchère et al. under the
assumption of a negative average order parameter in N,
\( \Delta_N/k_B \approx -160 \text{ mK} \). Moreover, some features of the
paramagnetic reentrant effect, such as nonlinearity, hys-
teresis, and dissipation, are in qualitative agreement with
their results.

At this point, a direct comparison of our experiment
with theory is not possible. The magnitude of \( \chi_{\text{para}} \)
of order 1, the characteristic dependence of the magnetic
susceptibility on \( T \) and \( L \), as described by Eq. [5] as well
as its field dependence remain to be obtained by the-
tory. More theoretical work and also more experiments
are necessary.

In summary, the paramagnetic reentrance phenomenon
in AgNb cylinders of high purity is a nonlinear effect of
anomalously strong magnitude. It shows strong devia-
tions from induced Meissner screening in the low-
temperature–low-field corner of the \( H-T \) phase diagram.
It displays dissipation, hysteresis, and long time re-
laxation behavior.

In the mesoscopic regime, the exponential temperature
dependence of the magnetic susceptibility with character-
istic temperature \( T^* \approx hV_F/2\pi k_B n L \) has the fingerprint
of several levels of quantum coherence along integer mul-
tiples \( n = 1, 2, 4 \) of the wire perimeter \( L \).

Paramagnetic reentrance has also been observed in
other NS materials. In our most recent experiments
we have found reentrant behavior in gold-niobium cylin-
ders. This has to be viewed in the light of expected su-
perconductivity in Au below \( T_c \approx 200 \mu \text{K} \). In consid-
eration of this, the origin of this puzzling paramagnetism
in mesoscopic NS cylinders is still an open question.

We wish to express our gratitude to R. Frassanito, M.
Nideröst, and P. Visani for their invaluable contributions
to earlier experimental work. We thank the group at the
ULT facility in Bayreuth for their help and support. We
acknowledge discussions with W. Belzig, G. Blatter, C.
Bruder, A. Fauchère, and Y. Imry. We acknowledge par-
tial support from the “Schweizerischer Nationalfonds zur
Förderung der Wissenschaftlichen Forschung” and the
“Bundesamt für Bildung und Wissenschaft” (EU Pro-
gram “Training and Mobility of Researchers”).

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