Reentrance of the induced diamagnetism in gold-niobium proximity cylinders

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We discuss the magnetic response of gold-niobium proximity cylinders in the temperature range $100 \mu K < T < 9 K$. The temperature dependence of the susceptibility in the diamagnetic regime is described well by the quasiclassical Eilenberger theory including an elastic mean-free path $\ell$. In the mesoscopic regime, the temperature dependence of the reentrant paramagnetic susceptibility $\chi_{\text{para}}(T) \propto \exp\left[-L/3\xi_N(T)\right]$ is governed by the dirty limit coherence length $\xi_N(T) = \sqrt{\frac{1}{3\varepsilon_0 N \ell_N}}$, with $\xi_N$ the clean limit coherence length obtained from breakdown field measurements and $\ell_N$ the measured mean-free path in the gold layer. At $T = 100 \mu K$, $\chi_{\text{para}}$ compensates about 1/5 of the induced diamagnetic susceptibility in gold.

The field of mesoscopic physics, including superconducting structures in proximity with normal metals, has attracted a wide interest, recently. In particular, experimental work on transport properties of mesoscopic proximity systems has been enabled by recent advances in nanostructure fabrication technology. In addition, extensive theoretical and experimental work on magnetic properties has led to the understanding of the high-temperature diamagnetic response of rather clean normal metal-superconductor (NS) proximity structures in the context of the quasiclassical Eilenberger theory as well as the magnetic breakdown transitions.

On the other hand, the paramagnetic reentrance phenomenon, discovered some years ago in silver-niobium cylinders, is still a matter of discussion. Recently, three theoretical works have addressed the origin of paramagnetic currents in NS systems from different points of view. Bruder and Imry consider non Andreev reflecting trajectories at the outer surface of NS proximity cylinders (glancing states), which give a paramagnetic correction to the Meissner susceptibility. This result has been subject to debate because of its small magnitude. The approach by Fauchère et al. assumes a net repulsive interaction in the noble metals. In their case, the $\pi$ shift of the order parameter then leads to spontaneous paramagnetic currents at the NS interface. In the most recent work by Maki and Haas, the noble metals copper, silver, and gold are assumed to show p-wave superconducting ordering below transition temperatures close to where the paramagnetic reentrance effect occurs. In their model, a quantized counter-current is generated in the periphery of the NS cylinders, compensating the Meissner effect. Unfortunately, neither of the present theories correctly explain the absolute value or the temperature dependence of the paramagnetic reentrant effect. A more detailed discussion can be found in Ref. 1.

In view of the most recent two theories, which depend on specific (assumed) properties of the normal metals, it is important to check the reentrant effect in NS junctions of different materials. Here, we report on the reentrance of the magnetic susceptibility of a gold-niobium sample, covering a large mesoscopic regime down to $\mu K$-temperatures. This new material combination gold-niobium, follows our previous investigations of the paramagnetic reentrant effect in AgNb and CuNb specimens superimposing on fully induced Meissner screening.

The sample reported here is an ensemble of cylindrical wires with a superconducting core of soft niobium ($RRR = 300$) concentrically embedded in a normal-metal matrix of gold. The total diameter was mechanically reduced by several steps of swaging and co-drawing to the final value $23 \mu m$, with average normal layer thickness $3.2 \mu m$. We chose not to anneal this sample, since gold dissolves in niobium as well as Nb in Au up to 5-10 percent for temperatures above $500^\circ C$. For this sample the value of the mean-free path $\ell_N \sim 0.3d_N$ was obtained from resistivity measurements. The detailed sample preparation was done as for the samples reported in Ref. 2.

A bundle of about 200 wires was placed directly inside the mixing chamber of a dilution refrigerator in contact with the liquid $^3$He-$^4$He solution. Using an rf-SQUID sensor, we measured inductively the temperature and field dependent ac-magnetic susceptibility as well as dc magnetization curves for $7 mK < T < 7 K$. For thermometry, the Curie-type magnetic susceptibility of the paramagnetic salt CMN (cerium magnesium nitrate) was measured, calibrated with Ge resistors.

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-SQUIV 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, 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. Thus, about 200 wires were mounted. Temperatures were measured with a pulsed NMR Pt thermometer.

In the following we report on the temperature dependent magnetic susceptibility of the gold–niobium sample 41AuNb [Fig. 1]. We show $\chi_{ac}(T)$ between $7 mK$ and $9 K$, measured in our dilution refrigerator with field amp-
Magnetic susceptibility $\chi_{ac}$ ($\bullet$), $\chi_{dc}$ ($\diamond$), and numerical fit with $\ell/d_N = 0.14$ (short-dashed line) of gold-niobium sample 41AuNb as a function of temperature ($H_{dc} = 0.2$ Oe). For comparison $\chi_{ac}$, $\chi_{dc}$ ($\times$), and numerical fit with $\ell/d_N = 0.55$ (long-dashed line) of silver-niobium sample 3AgNb.

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 NS interface. At lower temperatures, it develops almost total Meissner screening in the Au layer, as in comparable AgNb samples.

The numerically obtained curve with only one parameter $\ell$ fits the experimental data very well over the whole temperature range above the Andreev temperature $T_A = \hbar v_F/2\pi k_B d_N$. For sample 41AuNb it is $T_A = 530$ mK. The value of $\ell/d_N$ is 0.14, about a factor of two smaller than the value $\ell_N/d_N = 0.3$, obtained from resistivity measurements.

The fit is surprisingly good, if one takes into account the difference in geometry between experiment and theory, as well as the neglect of the boundary roughness in the theory and other imperfections inevitably present in the sample. The quasiclassical theory of the proximity effect with a finite mean-free path parameter $\ell$ due to a low concentration of elastic scatterers, is able to describe the linear susceptibility data of our relatively clean AuNb specimen as well as it was previously shown for AgNb and CuNb systems.

Supercooled ($\circ$) and superheated ($\bullet$) field as a function of temperature and exponential fits [see Eqn. 1] for sample 41AuNb. The different data correspond to the low-(high-)field transitions for effective gold thickness $d_N^{(1)} = 4.0 \mu$m ($d_N^{(2)} = 1.8 \mu$m) of the gold layer. The arrows indicate the direction of field changes.
For comparison, in Fig. 4 we also have plotted the temperature-dependent diamagnetic susceptibility of a silver-niobium sample described in Ref. 10 [sample 3AgNb, \( d_N = 5.5\, \mu m, T_A = 310\, mK \)] and its numerical fit with \( \ell / d_N = 0.55 \). A close examination of both curves shows that, the diamagnetic susceptibility of the gold-niobium sample develops not so steep as for the AgNb sample, due to a higher level of impurities. Moreover, as has been shown by the authors and in Fig. 1, there are considerable deviations of the experiment from the theoretical curves at temperatures \( T < T_A \), where the surface quality has a stronger influence, not accounted for in the theory.

We also have investigated the nonlinear-magnetic response in the low-temperature regime, where the induced diamagnetism shows approximately full Meissner screening. The magnetic breakdown of the induced superconductivity, a first-order phase transition, was measured for the gold-coated NS proximity sample. In Fig. 2(a), the isothermal nonlinear susceptibility as a function of magnetic field is shown for different temperatures between 7 mK and 400 mK. An isothermal magnetization curve for \( T = 7\, mK \) is displayed in Fig. 2(b).

The double transitions displayed in Fig. 2 are caused by the displacement of the niobium cores from the center of the cylinders during our drawing procedure of the niobium inside the soft gold matrix. The two nearly separated transitions (1) and (2) correspond to two different effective thicknesses of the gold layer \( d_N^{(1,2)} \). The supercooled and superheated fields \( H_{ac,sh} \) for each breakdown transition are shown as a function of temperature in Fig. 3. At temperatures well above 0.1\( T_A \), the experimental breakdown fields follow the exponential dependence

\[
H_{ac,sh}^{(1,2)}(T) \propto \frac{1}{\ell^{(1,2)}_N} \exp \left[ -d_N^{(1,2)}/\xi_N(T) \right], \tag{1}
\]

indicated in Fig. 3 as solid lines. This is similar to the theoretical clean limit expression as given in Ref. 4.

Experimentally we found

\[
\xi_N(T) = p \cdot \xi_T, \tag{2}
\]

with the clean limit thermal length in gold

\[
\xi_T = h v_F/2\pi k_B T = 1.7\, \mu m/T(K). \tag{3}
\]

For this gold-niobium sample 41AuNb, the value of the prefactor \( p \) is 0.5. Several other gold-niobium specimens showing paramagnetic reentrance have 0.4 < \( p < 0.7 \). The temperature dependence of Eqn. 3 with \( p < 1 \) is analogous to our findings in silver-niobium samples with 0.15 < \( \ell_N / d_N < 0.3 \) and copper samples indicating an elevated density of scattering centers with respect to the clean samples with \( p = 1 \).

In the linear magnetic susceptibility [Fig. 4], for sample 41AuNb below \( T_r \approx 40\, mK \) the signature of reentrance is observed, with the development of an additional paramagnetic susceptibility \( \chi_{para}(T) \), such that \( \chi_N(T) = \chi_{dia}(T) + \chi_{para}(T) \).

For this sample, the paramagnetic reentrance occurs at lower temperatures than for AgNb samples of similar sizes and shows a weaker increase as the temperature is reduced. In spite of the smaller value of \( \chi_{para} \) of the gold-niobium sample, the reentrant effect below \( \approx 500\, \mu K \) shows a strong temperature dependence with no signs of saturation down to the minimal temperature \( \approx 100\, \mu K \). The silver-niobium sample in Fig. 4 shows saturation of the magnetic susceptibility below \( \approx 400\, \mu K \), which could be intrinsic or lack of equilibrium. Indeed, long-time effects of the order of several days showing for the silver-niobium samples discussed in Ref. 10 have not been observed in sample 41AuNb.

Hysteresis effects which were very important in the silver-niobium samples described in Ref. 10 were not observed in the gold-niobium sample, possibly due to the very small value of \( \chi_{para} \) above 7 mK, where we could trace it on cooling and warming.

Fig. 4 shows the reentrant paramagnetic susceptibility below \( T_r \). The data exponentially increases as

\[
4\pi\chi_{para}(T) = A \exp \left( -\sqrt{T/T_0} \right), \tag{4}
\]

with characteristic temperature \( T_0 = 1\, mK \) and prefactor \( A = 0.23 \). The value of the characteristic temperature can be connected to an energy through \( k_B T_0 = 4.7 h v_F l_N/6\pi L^2 \), with \( l_N \) the measured mean-free path and \( L = 72\, \mu m \) the wire perimeter.

Alternatively, we use the experimental coherence length of the Andreev pairs \( \xi_N = p\xi_T \) in Au, obtained from our breakdown field measurements, in the expression for the dirty limit coherence length \( \xi_N^d = \sqrt{1/3\xi_N^2\ell_N} \). Thus, we find for the temperature dependence of the paramagnetic reentrant susceptibility

\[
\chi_{para}(T) = 4\pi A \exp \left( -\sqrt{T/T_0} \right), \tag{5}
\]

with characteristic temperature \( T_0 = 1\, mK \) and prefactor \( A = 0.23 \). The value of the characteristic temperature can be connected to an energy through \( k_B T_0 = 4.7 h v_F l_N/6\pi L^2 \), with \( l_N \) the measured mean-free path and \( L = 72\, \mu m \) the wire perimeter.
\[4\pi \chi_{\text{para}} = A \exp \left( -\frac{L}{3\xi_N^d(T)} \right).\] (5)

The dirty-limit coherence length \(\xi_N^d(T)\) in Eqn. 3 again indicates the stronger level of impurities in the gold-niobium sample 41AuNb as compared to the silver-niobium samples discussed in Ref. 10. Due to the short mean-free path \(\ell_N\) the induced diamagnetism is only slightly weaker as compared to sample 3AgNb, whereas the paramagnetic reentrant susceptibility is in the dirty regime. \(\chi_{\text{para}}\) develops below a lower characteristic temperature \(T_r\) and is by a factor of five smaller than for sample 3AgNb. From these results it is evident that, impurities have a much stronger influence on the paramagnetic reentrance phenomenon than on the proximity effect.

In summary, we have found the paramagnetic reentrance phenomenon in gold-coated niobium proximity cylinders. For these NS samples, the absolute value of the paramagnetic reentrant susceptibility is considerably smaller than in clean silver-niobium specimens, showing a stronger influence of scattering centers, the quality of the NS interface, as well as the free surface. The temperature dependence of \(\chi_{\text{para}}\) is governed by the dirty limit coherence length, which becomes of the order of the wire perimeter.

The paramagnetic reentrant behavior of gold–niobium cylinders has to be viewed in light of the expected superconductivity in Au below \(T_c \approx 200 \mu\text{K}\) for a very pure gold sample, as well as the theoretical assumptions for this element.

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