## Novel Reentrant Effect in the Proximity-Induced Superconducting Behavior of Silver

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A new reentrant behavior of the ac susceptibility in proximity-induced superconductivity of silver has been discovered. This is observed when the induced coherence length becomes comparable to the size of the specimen. The effect is destroyed by applying a magnetic field or by raising the temperature. This phenomenon points to the existence of a new form of quantum coherence effect in clean proximityinduced superconductors.

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Lately, bulk superconductivity has been induced by the proximity effect at millikelvin temperatures in the otherwise normal metals copper and silver for thicknesses up to 0.1 mm. The magnetic properties of these proximity systems have been discussed recently.<sup>1</sup>

In this Letter we present data on a novel reentrant effect on Ag-Nb proximity-effect samples with a bulk mean free path  $l_N^{\text{bulk}}$  very long compared to the thickness  $d_N$  of the silver layer. Typical values for  $l_N^{\text{bulk}}$  were around 20  $\mu$ m.

The specimens consisted of wires with one filament of Nb embedded in a high-purity Ag matrix. Specimens with different values for the thicknesses  $d_N$  and  $d_S$  of the normal and superconducting metals were obtained by a codrawing procedure. The samples were mounted inside the mixing chamber of a dilution refrigerator in direct contact with the <sup>3</sup>He-<sup>4</sup>He solution. Isothermal dc magnetization curves and ac susceptibility as a function of field were measured with a noncommercial SQUID magnetometer. The real in-phase component ( $\chi'$ ) and the imaginary out-of-phase component ( $\chi''$ ) of the ac susceptibility were measured using frequencies between 16 and 160 Hz with typical measuring ac fields smaller than 33 mOe peak to peak. The residual field in the cryostat is approximately 2 mOe.

Figure 1 shows the measured components of the zerofield ac susceptibility  $\chi'$  and  $\chi''$  as functions of temperature, for the sample with the smallest value of the normal-metal thickness  $d_N = 3.3 \ \mu m$ . The logarithmic horizontal scale has been used to display better the lowtemperature region; the vertical scale for  $\chi'$  has been calibrated to give a value of  $\chi'=0$  for a full diamagnetic Nb core and  $\chi'=-1$  for complete screening in silver. As the temperature is lowered below  $T_c$  of the Nb core, leakage of Cooper pairs into the normal-metal (Ag) contributes progressively to screen the silver from the measuring ac field, giving rise to an increase in the absolute value of the diamagnetic response  $\chi'$ , while  $\chi''$ remains practically constant. This screening induced by the proximity effect has been observed by many authors both in copper<sup>2-4</sup> and in silver.<sup>5</sup>

The new striking effect appears by further cooling the

sample below the temperature  $T_{\min}$  for which complete screening of the sample is observed. For  $T < T_{min}$ , the  $\chi'$ signal does not saturate at  $\chi' = -1$  as observed in thicker specimens, but returns back toward smaller absolute values. At our lowest temperature T=4 mK, it reaches the level corresponding to the complete shielding of Nb, within the uncertainty of the  $\chi'$  measurement. The outof-phase component  $\chi''$ , which is practically constant down to  $T = T_{min}$ , dramatically changes below  $T_{min}$ . Both components of the ac susceptibility have also been recorded as a function of frequency at all temperatures. The frequency dependence of  $\chi'$  and  $\chi''$  (in the frequency range 16 < v < 160 Hz) is negligible. We have failed, however, to detect distinct features of the reentrant behavior by measuring the initial slope of dc magnetization curves. This is presumably due to the lower sensitivity of the dc measuring system with respect to the phasesensitive technique employed in the ac susceptibility measurements.

The size dependence of the reentrant behavior is

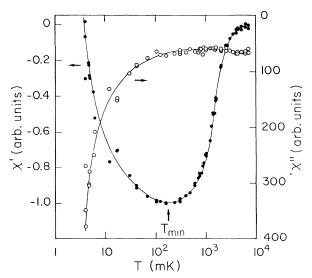


FIG. 1. In-phase (•) and out-of-phase ( $\bigcirc$ ) components of ac susceptibility as a function of temperature for a Ag-Nb sample with  $d_N = 3.3 \ \mu$ m. The lines are guides to the eye.

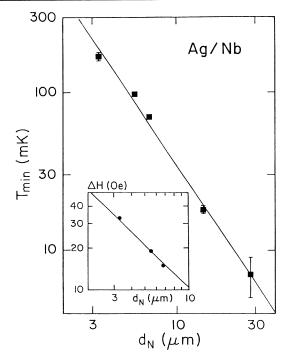


FIG. 2. Double-logarithmic plot of  $T_{\min}$  as a function of normal-metal thickness  $d_N$ . Inset: The quantity  $\Delta H$ , as defined in the text, as a function of  $d_N$ .

shown in Fig. 2, where the experimental values of  $T_{min}$  for five different samples have been plotted as a function of the thickness of the normal-metal  $d_N$ . The data are well described by a power law of the following type:

$$T_{\min} \propto (d_N)^{-\beta} \tag{1}$$

with  $\beta$  around 1.5.

Undoubtedly a new effect develops as T is reduced, which decreases the maximum diamagnetic shielding observed at  $T = T_{min}$ . We can investigate the temperature and field dependence of this new phenomenon by considering the susceptibility  $\chi'$  as a function of T and H for  $T < T_{min}$ . The change in screening below  $T_{min}$  is given by  $\Delta \chi' = \chi'(T) - \chi'(T_{min})$  at H = 0. In Fig. 3 this difference has been plotted as a function of temperature for  $T < T_{min}$  for a sample with  $d_N = 5.5 \ \mu$ m. The fitted curve corresponds to

$$\Delta \chi' = A \exp(-T/T^0) \tag{2}$$

with A = 0.206 (arbitrary units) and  $T^0 = 15.7$  mK.

The result of this type of analysis extended to the other samples is given in the inset of Fig. 3 where  $T^0$  and Ahave been plotted as a function of the normal-metal thickness  $d_N$ . The characteristic temperature  $T^0$  and the coefficient A depend on the size of the sample as

$$T^0 d_N \approx \text{const} = 0.081 \ \mu \text{m K}$$
,  
 $A d_N^2 \approx \text{const} = 6.5 \ \mu \text{m}^2 \text{ arbitrary-units}$ .
(3)

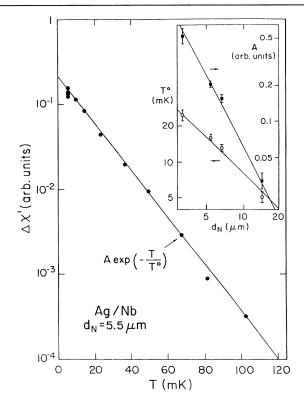


FIG. 3.  $\Delta \chi' = \chi'(T) - \chi'(T_{\min})$  as a function of temperature for a Ag-Nb sample with  $d_N = 5.5 \,\mu$ m. Inset: The characteristic temperature  $T^0$  (O) and the coefficient A (**n**), for four samples, as a function of normal-metal thickness  $d_N$ .

Measurements of the in-phase component  $\chi'$  of the susceptibility have also been made for each sample as a function of an external dc magnetic field applied parallel to the longitudinal axis of the sample as described elsewhere.<sup>1</sup> An example of the experimental results taken at T=4 mK is given in Fig. 4 for the thinnest sample. As the external field is increased,  $\chi'$  changes, returning towards its value at  $T = T_{\min}$  and H = 0 corresponding to complete diamagnetic shielding. More precisely, for the specimen with  $d_N = 3.3 \ \mu m$  only 70% of that value is reached at  $H \approx 17$  Oe. On further increasing the field, the breakdown transition occurs at  $H_b \approx 20$  Oe, as already seen in other proximity-effect superconductors.<sup>1</sup> In the inset of Fig. 4 we give  $\chi'$  for H < 20 Oe and both field polarities. The hysteresis and slight asymmetry of the data might be due to some trapped flux. The quantity  $\Delta H$  indicated in the inset of Fig. 4 is also strongly size dependent and a plot of the extrapolated value of  $\Delta H$  for  $T \rightarrow 0$  as a function of  $d_N$  is given in the inset of Fig. 2 for the three thinnest samples only. Clearly  $\Delta H$  is inversely proportional to  $d_N$ .

The interpretation of these results is not trivial. Impurities and spurious effects are ruled out by the data shown above which clearly demonstrate that the reentrant effect is intimately related to the sample dimensions.

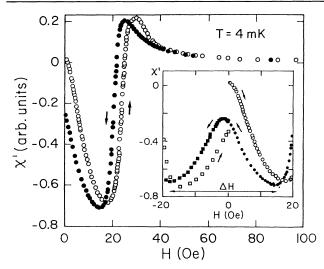


FIG. 4. In-phase component of the ac susceptibility as a function of external dc magnetic field for a sample with  $d_N = 3.3 \ \mu m$ . Inset: The susceptibility  $\chi'$  for both magnetic-field polarities in the interval -20 < H < 20 Oe. The arrows indicate the field sweep direction.

It seems clear that the reentrant behavior described here is another manifestation of long-range quantum coherence. In proximity-effect samples the Cooper-pair penetration length  $K_N^{-1}$ , which gives the scale of variation of the proximity-induced order parameter in silver, increases by lowering the temperature as  $K_N^{-1} = \alpha/T$ , with  $\alpha$  values for the five samples between 1.3 and 1.7  $\mu$ mK as obtained from breakdown-field data.<sup>1,5</sup> This means that by lowering the temperature it is easy to enter into a regime where the pair-penetration length  $K_N^{-1}$  becomes comparable to or even bigger than the size of the samples. For example, at T=10 mK we already have values of  $K_N^{-1}$  as big as 0.15 mm.

In the case of normal metals a number of well-known interesting effects<sup>6</sup> appear when the coherence of the single-particle wave function is maintained across the sample. Critical behaviors are also expected for superconductors once the coherence length  $\xi$  is comparable to the sample dimensions. As pointed out for the first time by de Gennes,<sup>7</sup> the response of an isolated superconducting loop depends on the ratio between the radius of the loop R and the coherence length  $\xi$ . He showed that, if the ratio  $R/\xi$  is smaller than  $\frac{1}{2}$ , values of critical flux  $\Phi_c$ given by  $\cos(2\pi\Phi_c/\Phi_0) = \cos(2\pi R/\xi)$  are delimiting an interval around  $\Phi = (n + \frac{1}{2})\Phi_0$  where superconductivity cannot exist even at T=0; here n is an integer and  $\Phi_0 = h/2e$  is the flux quantum. The interval around  $\Phi = (n + \frac{1}{2})\Phi_0$  where diamagnetism vanishes depends on the value of the ratio  $R/\xi$ ; for smaller values of this ratio

TABLE I. Values of normal-metal thickness  $d_N$  and Nb core diameter  $d_S$ . The ratio between Cooper-pair penetration length  $K_N^{-1}$  and  $d_N$  as well as the one between  $K_N^{-1}$  and mean radius R are both given for  $T = T_{min}$ .

$d_N$ ( $\mu$ m)	<i>ds</i> (µm)	$K_N^{-1}/d_N$	$K_N^{-1}/R$
3.3	16.4	2.9	1.0
5.5	30	2.8	0.9
6.75	36.5	3.2	1.0
14.5	71	5.0	1.7
28	135	7.4	2.5

the interval becomes bigger. Unfortunately, for an intrinsic superconductor the condition described above is fulfilled only in extremely small samples, so that inductive measurements become unaccessible.<sup>8</sup>

In Table I, we give values of  $d_N$ ,  $d_S$ ,  $K_N^{-1}/d_N$  at  $T = T_{\min}$ , and  $K_N^{-1}/R$  at  $T = T_{\min}$  for the five specimens studied here. R is the mean radius of the silver cylindrical shell. It is surely interesting to notice that for the three thinnest specimens at  $T = T_{\min}$  the relation  $K_N^{-1} \approx 3d_N = R$  is fulfilled.

Although de Gennes' predictions for a single loop cannot be directly compared to the results obtained with our proximity-effect sample geometry, we believe that the underlying idea of the vanishing of the diamagnetism when the dimensions of the loop are comparable to the coherence length is closely related to the reentrant effect we report here.

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<sup>3</sup>A. C. Mota, D. Marek, and J. C. Weber, Helv. Phys. Acta 55, 647 (1982).

<sup>4</sup>Th. Bergmann, K. H. Kuhl, B. Schröder, M. Jutzler, and F. Pobell, J. Low Temp. Phys. **66**, 209 (1987).

<sup>5</sup>A. C. Mota, P. Visani, and A. Pollini, Jpn. J. Appl. Phys. **26**, 1693 (1987).

<sup>6</sup>S. Washburn and R. A. Webb, Adv. Phys. 35, 375 (1986).

<sup>7</sup>P. G. de Gennes, C. R. Acad. Sci. Ser. 2 **292**, 9 (1981); **292**, 279 (1981).

<sup>8</sup>P. Santhanam, C. P. Umbach, and C. C. Chi, Phys. Rev. B **40**, 11 392 (1989).

<sup>&</sup>lt;sup>1</sup>A. C. Mota, P. Visani, and A. Pollini, J. Low Temp. Phys. **76**, 465 (1989).

<sup>&</sup>lt;sup>2</sup>Y. Oda and H. Nagano, J. Phys. Soc. Jpn. 44, 2007 (1978).