

# Diamagnetic response of cylindrical normal-metal–superconductor proximity structures with low concentration of scattering centers

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We have investigated the diamagnetic response of composite NS proximity wires, consisting of a clean silver or copper coating, in good electrical contact to a superconducting niobium or tantalum core. The samples show strong induced diamagnetism in the normal layer, resulting in a nearly complete Meissner screening at low temperatures. The temperature dependence of the linear diamagnetic susceptibility data is successfully described by the quasiclassical Eilenberger theory including elastic scattering characterized by a mean-free path  $l$ . Using the mean-free path as the only fit parameter we found values of  $l$  in the range 0.1–1 of the normal metal-layer thickness  $d_N$ , which are in rough agreement with the ones obtained from residual resistivity measurements. The fits are satisfactory over the whole temperature range between 5 mK and 7 K for values of  $d_N$  varying between 1.6 and 30  $\mu\text{m}$ . Although a finite mean-free path is necessary to correctly describe the temperature dependence of the linear-response diamagnetic susceptibility, the measured breakdown fields in the nonlinear regime follow the temperature and thickness dependence given by the clean limit theory. However, there is a discrepancy in the absolute values. We argue that in order to reach quantitative agreement one needs to take into account the mean-free path from the fits of the linear response. [S0163-1829(99)08809-8]

## I. INTRODUCTION

A normal metal (N) in good electrical contact with a superconductor (S) exhibits superconducting properties as the temperature is reduced. First experiments on the proximity effect were reported by Holm and Meissner,<sup>1</sup> who observed zero resistance between SNS pressed contacts. Since then, many investigations on proximity effects have been carried out.<sup>2</sup> Recently, it has received a revived interest.<sup>3</sup> Particularly, experiments on the magnetic response have demonstrated nontrivial screening properties, showing hysteretic magnetic breakdown at finite external fields<sup>4–6</sup> as well as a presently unexplained reentrant effect at low temperatures.<sup>7</sup>

First theoretical studies on the proximity effect were carried out by Cooper.<sup>8</sup> For a diffusive proximity system, the diamagnetic susceptibility in the linear regime was investigated by the Orsay Group on Superconductivity in the framework of the Ginzburg–Landau theory.<sup>9</sup> In this limit, it was found that the induced diamagnetic susceptibility in N depends on temperature approximately as  $\chi_N \propto T^{-1/2}$ . This was confirmed experimentally by Oda and Nagano.<sup>10</sup> In spite of that, the magnetic properties of proximity samples discussed in Refs. 4 and 6 could not be explained by the Ginzburg–Landau theory, since they show a much stronger temperature dependence of the diamagnetic susceptibility.

The clean limit, which is defined for the elastic mean-free path  $l \rightarrow \infty$ , was first studied by Zaikin<sup>11</sup> for a finite system of ideal geometry with the help of the quasiclassical theory. He predicted that, since in this case the current-field relation is completely nonlocal, the current in N is spatially constant and depends on the vector potential integrated over the whole normal metal. As a consequence, the magnetic flux is screened linearly over the normal layer thickness  $d_N$  and for

$T \rightarrow 0$  the susceptibility can reach only  $\frac{3}{4}$  of that of a perfect diamagnet. For  $T \gg T_A$  he found  $\chi_N \propto \exp(-2T/T_A)$ , with the Andreev temperature  $T_A = \hbar v_F / 2\pi k_B d_N$ . This mesoscopic temperature originates from the energy of the lowest Andreev bound state.<sup>12</sup> In the ballistic case, all properties of the proximity effect are determined by these bound states, which are coherent superpositions of electron and hole waves between consecutive Andreev and normal reflections.

Within the framework of the quasiclassical theory, Belzig, Bruder, and Schön<sup>13</sup> studied recently the magnetic response of a proximity coupled NS sandwich in the two limits, clean and dirty. They obtained numerical solutions of the corresponding equations for a wide range of temperature, magnetic field, and layer thickness. Furthermore, they tried a fit of the susceptibility data of one AgNb specimen with a relatively small mean-free path (specimen 1 AgNb in this paper) within their dirty limit results. The fit was only successful at very low temperatures, but could not reproduce the high-temperature data. The diamagnetic screening of the dirty limit was too big as compared to the experiment. Moreover, the temperature dependence of the susceptibility of NS proximity specimens with the longest mean-free paths could not be explained within the clean limit theory. This limit gives a completely different temperature dependence than the one observed in the experiments. In addition, data at low temperatures reach almost perfect diamagnetic screening, whereas in the clean limit it should not exceed 75% of full screening. To close the gap between these two limits the linear-magnetic response for arbitrary impurity concentrations was theoretically studied in Ref. 14. We show in this paper that these results can satisfactorily describe the experimental data.

The theory of the nonlinear response was addressed in

Refs. 9, 13, and 15. A proximity NS sandwich with a finite normal layer thickness  $d_N$  at sufficiently low temperatures undergoes a first-order phase transition both in the clean and in the dirty limit. At a breakdown field a jump in the magnetization occurs. In the clean limit the temperature dependence of the breakdown field is  $H_b(T) \propto \exp(-T/T_A)$ . We show in this paper, that the clean-limit result<sup>15</sup> agrees well with our experiments on  $H_b$ , which follow the same temperature dependence in relatively clean specimens ( $l/d_N \approx 0.4 - 0.8$ ), as well as the  $1/d_N$  dependence in the prefactor of  $H_b$  as found in previous experiments.<sup>6</sup> However, this is not the case for the shape of the magnetization curves and, as mentioned before, for the temperature dependence of the linear susceptibility.

We have fabricated a new set of CuNb and AgNb specimens similar to the ones reported in Ref. 6. The specimens were produced with an optimized annealing procedure in order to achieve very high mean-free paths. The ratio of the thickness of the normal layer to the radius of the superconductor of the composite wires,  $d_N/r_S$ , was also varied to investigate comparable samples with normal layers reaching from almost flat to rather curved cylindrical geometry. Thus, the influence of the NS geometry on the proximity effect was investigated. In this paper, we discuss these new samples together with older ones,<sup>6</sup> covering a wide range of parameters, and fit their diamagnetic response with the help of the quasiclassical theory in an intermediate impurity regime between the clean and dirty limit. Here, we will not consider the reentrant effect found by Mota and co-workers<sup>7</sup> but only note that a recent theoretical study has addressed this new phenomenon.<sup>16</sup> For the nonlinear response we compare the temperature dependent breakdown fields with the quasiclassical clean-limit result.

This paper is organized as follows. In Sec. II, we describe some theoretical aspects of the magnetic response. In Sec. III, we describe the sample preparation and the measurement apparatus. In Sec. IV, the experimental results on the linear magnetic response are presented and the data are fitted using the theory from Sec. II. The experimental results on the nonlinear response are addressed in Sec. V. Finally, we draw some conclusions.

## II. THEORETICAL ASPECTS

Within the quasiclassical theory of superconductivity<sup>17-19</sup> the magnetic response of a planar NS-proximity structure was investigated for arbitrary impurity concentrations by Belzig, Bruder, and Fauchère.<sup>14</sup> Below we summarize some basic results of this paper. The theoretical system consists of a semi-infinite superconductor in perfect contact with a normal metal of thickness  $d_N$  and specular reflecting outer surface. The pair potential  $\Delta$  is taken to be constant in the superconductor and to be zero in the normal metal. The mean-free path  $l$  and the Fermi velocity  $v_F$  are assumed to be the same throughout the system.

In Ref. 14 a variety of regimes are discussed, where the proximity effect is different from the previously studied clean and dirty limits. The regimes differ by the relative magnitudes of the thermal length  $\xi_T = \hbar v_F / 2\pi k_B T$ , the mean-free path  $l$ , and the thickness  $d_N$ , an additional relevant length scale, which has to be taken into account. In the

case of a finite normal layer thickness  $d_N$ , the clean and the dirty limit are restricted to much smaller parameter regions than previously believed. The dirty limit holds for  $l \ll \xi_T, d_N$  only, if also the mean-free path  $l \ll \xi_0$ . Here,  $\xi_0 \sim v_F / \Delta$  is the coherence length in the superconductor. On the other hand, the regime  $l < \xi_T$  belongs to the ballistic regime, if  $l > d_N$ .

To treat the screening problem, a general linear-response formula was derived, which yields a nonlocal current-field relation in terms of the zero-field Green's functions. These functions are obtained from the solution of the Eilenberger equation including an impurity selfenergy. In general this has to be done numerically, since analytical expressions can be derived only in the clean and in the dirty limit. Finally, the Maxwell equations have to be solved.

The chosen geometry is such that the NS interface lies in the  $y$ - $z$  plane. Then the current in the normal metal has the form

$$j_y(x) = - \int_0^{d_N} K(x, x') A_y(x') dx', \quad (1)$$

where  $A_y$  is the vector potential in transverse gauge, if the magnetic field is applied in  $z$  direction. The kernel  $K(x, x')$  contains an exponential dependence on the distance  $|x - x'|$ . The range of the kernel is given by  $l$  in most of the temperature and impurity regimes. For  $l > \xi_T$  and  $l \ll d_N \exp(2d_N/\xi_T)$  that is, at high temperatures, it is given by  $\xi_T$ . It has thus a strong temperature dependence that has to be contrasted to the case of a bulk superconductor, where the range of the Pippard kernel is only weakly temperature dependent.

The prefactor of the exponential in the kernel, which in general depends on coordinates, is related to the local superfluid density. It introduces an additional length scale in the problem, the field penetration depth  $\lambda(x, T) = \lambda_N f(x, T)$ , where  $\lambda_N = (4\pi e^2 n/m)^{-1/2}$  is a London-like length in the normal metal and  $f(x, T)$  is a function of temperature and position. The interplay between the range of the kernel and the superfluid density determines whether the nonlocal form of the current-field relation is important. This has strong consequences on the screening properties, especially in the case  $l > d_N$ , which is discussed in detail in Ref. 14.

In Fig. 1, we reproduce some numerical results<sup>14</sup> of the screening fraction  $\rho/d_N = -4\pi\chi_N$  in a normal metal layer for  $\lambda_N = 0.003d_N$ . Five curves as a function of temperature, normalized to the Andreev temperature, are given for different mean-free paths  $l$ .

For the clean limit,  $l \rightarrow \infty$ , at zero temperature, screening can only reach  $\frac{3}{4}$  of  $-1/4\pi$ . This is due to the infinite range of the kernel, or in other words, the complete nonlocality of the current-field relation. At a temperature  $T \approx 5T_A$ , screening is exponentially reduced by thermal occupation of the Andreev levels,<sup>20</sup> and at higher temperatures screening is almost negligible.

For  $l = 10d_N$  and  $l = d_N$  screening is enhanced in comparison to the clean limit at low and high temperatures. At low temperatures screening is more effective for  $d_N \leq l < \infty$ , since the range of the kernel is given by finite  $l$ . At high temperatures, where the range of the kernel is  $\xi_T$ , screening is more

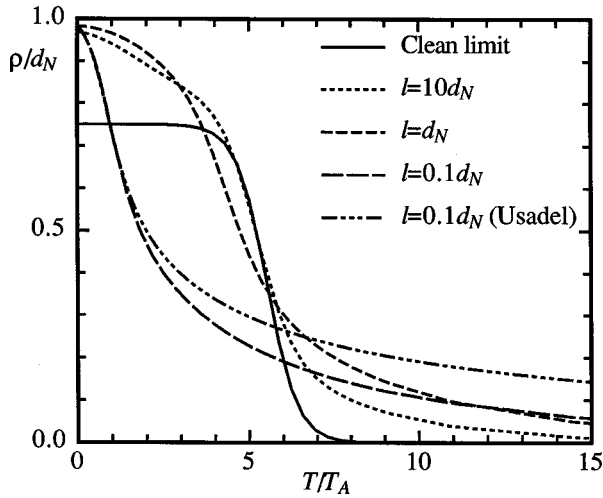


FIG. 1. Numerical results of the screening fraction of an NS proximity slab for  $\lambda_N = 0.003d_N$  and different impurity regimes as functions of  $T/T_A$ . The clean limit is given by a solid line and the Usadel theory result for  $l = 0.1d_N$  is given by a dash-dotted line. For the intermediate impurity regime three cases are given:  $l = 10d_N$ ,  $l = d_N$ , and  $l = 0.1d_N$ .

effective than in the clean limit but still nonlocal. For  $l = 0.1d_N$ , already at  $T \sim T_A$  screening is considerably reduced, since the superfluid density is suppressed because the mean-free path is smaller than  $d_N$ .

In the diffusive regime, it can be shown that for  $l^2 < \lambda_N d_N$ , the nonlocal relation is reduced to the local current-field relation of the Usadel theory.<sup>21</sup> In particular, for the case  $\lambda_N = 0.003d_N$  corresponding approximately to the experiments discussed in this paper, the condition  $l \ll d_N$  has to be met. Indeed, in Fig. 1 we illustrate the non-negligible difference in screening even for the case  $l = 0.1d_N$ , between the local Usadel result and the nonlocal approach with elastic scattering described in Ref. 14. For  $T \lesssim 1.5T_A$  the Usadel result exhibits a much more pronounced screening tail.

After these recent results, covering a wide range between the clean and the dirty limits, we have now the possibility to analyze the magnetic response of our relatively clean NS proximity specimens. To compare the experiments with the theory we have used the full form (1) valid for all impurity concentrations and solved the screening problem for each specimen numerically.

In these theoretical considerations the effect of a rough normal metal-vacuum boundary has been neglected completely. On the other hand, at high enough temperatures  $T \gg T_A$ , where the induced diamagnetism strongly decreases, the influence of surface scattering does not play an important role, since in this temperature regime the superfluid density at the outer boundary is exponentially suppressed. Screening in this regime can be only due to bulk scattering centers and is insensitive to the quality of the surface.

The quasiclassical theory for the nonlinear response in the clean limit was discussed in detail by Fauchère and Blatter.<sup>15</sup> They derived analytical expressions for the temperature dependent breakdown field  $H_b(T)$  of a clean normal-metal slab of finite thickness in proximity with a bulk superconductor in the two limits  $T \rightarrow 0$  and  $T \gg T_A$ . The breakdown

field  $H_b$  was determined with a Maxwell construction from the magnetization at the bistable regime, where two different values of the free energy coexist, characterizing a diamagnetic and a field penetration phase. The spinodals of that bistable regime have already been determined in the clean and in the dirty limit by Belzig, Bruder, and Schön<sup>13</sup> numerically. They represent the boundaries of superheating and supercooling in the first-order phase transition. The thermodynamical magnetic breakdown field lies in between the spinodals. It was determined by the intersection of the two asymptotics of the free energy.

For  $T \rightarrow 0$  the breakdown field saturates at<sup>15</sup>

$$H_b(0) \approx \frac{1}{6} \frac{\Phi_0}{\lambda_N d_N}, \quad (2)$$

whereas for  $T \gg T_A$  it decays as<sup>15</sup>

$$H_b(T) \approx \frac{\sqrt{2}}{\pi} \frac{\Phi_0}{\lambda_N d_N} e^{-d_N/\xi_T}. \quad (3)$$

The temperature dependence is a simple exponential with the exponent  $d_N/\xi_T = T/T_A$ . The prefactor of the breakdown field was found to scale inversely proportional to the normal layer thickness. Both features are in agreement with previous experimental results on  $H_b$  in relatively clean AgNb specimens.<sup>6</sup>

In the  $H$ - $T$  phase diagram a critical temperature  $T_{\text{crit}}$  was found, below which the first-order phase transition is observable. Above  $T_{\text{crit}}$  a continuous and reversible crossover between the diamagnetic and field-penetration regime occurs.

Since the breakdown field depends only on thermodynamical considerations, it is expected to be less sensitive to the nonlocality of the current-field relation than the linear susceptibility. A cylindrical geometry and a barrier at the NS interface is expected to change the breakdown field only quantitatively.<sup>15</sup> On the other hand, the influence of scattering centers is reflected in the shape of the magnetization curves.<sup>13</sup> At present, no theoretical results for the breakdown field or magnetization curves exist for intermediate impurity concentrations.

### III. SAMPLE PREPARATION AND EXPERIMENTAL SETUP

The samples we investigated are bundles of cylindrical wires with a superconducting core of niobium or tantalum concentrically embedded in a normal metal matrix of copper or silver. The normal metal starting materials are of purity 4N to 6N with negligible contribution of magnetic impurities. The purity of the niobium and tantalum starting materials was the highest accessible ( $RRR \approx 300$ ), resulting in a good ductibility. We assembled the samples by placing in a close fit, a rod of the S material inside a hollow cylinder of the N material, with typical outer diameter of about 20 mm. The cleanliness of the internal N tube surface and the S rod surface is very important. The surfaces of the initial metal pieces were cleaned in acid and mechanically smoothed with a special tool in order to provide a good metallic contact. All

TABLE I. Table of sample parameters. The measured value  $l_N/d_N$  and the fitting parameter  $l/d_N$  are given. In the last column the saturated superheated field at the lowest temperature  $H_{sh}^{\text{sat}}$  is shown.

Sample	$d_N$ [ $\mu\text{m}$ ]	Ratio $d_N/r_S$	$T_{\text{ann}}$ [ $^{\circ}\text{C}$ ]	$l_N$ [ $\mu\text{m}$ ]	$T_A$ [mK]	$\lambda_N$ [nm]	$l_N/d_N$	$l/d_N$	$H_{sh}^{\text{sat}}$ [Oe]
1AgNb	14.5	0.4	not	1.8	120	22	0.12	0.15	1.6
2CuNb	15.3	1.0	not	0.9	120	18.3	0.06	0.025	0.4
3AgNb	5.5	0.4	700	4.3	310	22	0.78	0.65	15.2
4AgNb	6.8	0.4	700	5.1	250	22	0.75	0.75	12.9
5AgNb	3.3	0.4	800	1.6	520	22	0.49	0.55	24.7
6AgNb	28.0	0.4	550	9.0	61	22	0.32	0.25	2.1
7CuNb	5.3	1.0	not	0.9	360	18.3	0.18	0.08	5.9
12AgNb	3.6	0.4	not	1.8	480	22	0.5	0.26	15.4
14CuTa	5.0	1.0	not	1.0	380	18.3	0.2	0.07	7.0
16AgNb	3.3	0.4	800	1.7	520	22	0.5	0.35	24.5
19CuTa	3.8	1.0	600	2.5	510	18.3	0.67	0.13	12.9
20AgNb	2.7	0.4	not	1.4	630	22	0.53	0.24	20.2
21AgNb	3.6	0.4	800	1.0	480	22	0.28	0.75	15.8
23CuNb	5.7	0.2	not	1.4	340	18.3	0.24	0.11	8.4
24CuNb	2.5	0.2	not	0.9	760	18.3	0.35	0.15	23.9
25CuNb	1.6	0.2	not	0.5	1200	18.3	0.27	0.19	40.5
28AgNb	14.5	0.4	700	5.6	120	22	0.39	0.45	4.9
33AgNb	7.7	1.0	700	5.3	220	22	0.7	0.35	10.2
34AgNb	3.1	0.4	700	4.0	540	22	1.27	0.55	26.2
35AgNb	2.8	0.4	670	3.6	620	22	1.3	0.55	30.8
36AgNb	5.1	1.0	670	5.4	330	22	1.05	0.4	14.1
37AgNb	4.2	0.4	670	3.5	410	22	0.84	0.45	18.9
38AgNb	3.8	0.4	not	1.2	450	22	0.32	0.18	10.6

this was done in an argon atmosphere. The NS cylinder was then embedded in a Cu mantle to protect the sample itself from contamination. After assembly, the diameter was reduced mechanically by several steps of swagging and codrawing<sup>22</sup> down to a total diameter of several hundred microns.

After etching away the Cu protection, the diameter of the NS specimens was further reduced by codrawing the wires through several diamond dies to final values between 15 and 190  $\mu\text{m}$ . The extreme size reduction (by factors of up to 1000) by codrawing resulted in a great enhancement of the quality of the NS interface for electronic transmission. The use of high purity starting materials guaranteed a high electronic mean-free path. Some of the samples were annealed after the last drawing in an atmosphere of argon at a temperature around 650  $^{\circ}\text{C}$  for about one hour to remove the effect of cold working in the normal metal. For that purpose the wire of selected size was wound on a small Ag plate. As a consequence of the optimized annealing procedure, values of  $l_N \geq d_N$  could be achieved. The mean-free paths were obtained from resistivity measurements along the wires, performed at a temperature  $T = 10$  K with a four-point method. The thickness of the normal metal layer as well as the surface quality of the wires were determined with the help of scanning electron micrographs. Several samples were prepared in order to cover a large range of parameter space. We fabricated AgNb samples with a ratio  $d_N/r_S$  of 0.4 or 1. The layer thicknesses ranged from  $d_N = 2.8 \mu\text{m}$  to  $d_N = 28 \mu\text{m}$  with measured mean-free paths ranging from  $l_N/d_N = 0.12$  to

$l_N/d_N = 1.3$ . Moreover, we prepared CuNb and CuTa samples, with varying ratios of  $N$  layer thickness to  $S$  core radius, and mean-free path  $l_N$ . This gave us the opportunity to apply the theory described in Sec. II in a wide range of samples. The values of the sample parameters are listed in Table I.

As a last step, the wires were glued with GE7031 varnish and thus, electrically insulated. After drying, they were removed from the silver support, cut to a length of typically 3 to 5 mm and rolled together forming a bundle of 200 to 800 wires. The wire bundle was then placed directly inside the mixing chamber of a dilution refrigerator in contact with the liquid  $^3\text{He}$ - $^4\text{He}$  solution, mounted parallel to the coil axis of the magnetic ac and dc fields.

The ac magnetic susceptibility was measured at temperatures between 5 mK and 7 K using an rf-superconducting quantum interference device (SQUID) sensor, inductively coupled via a dc-flux transformer to the sample. A certain fraction of the voltage applied to the primary ac coil was also mutually fed into the flux transformer loop, using the SQUID as a null detector. This allowed us to measure the susceptibility of the samples with a relative precision of about  $10^{-4}$ . Typical ac amplitudes  $H_{\text{ac}}$  are between 0.06 and 33 mOe, with frequencies 16, 32, 80, and 160 Hz. The temperature was measured through the Curie-type magnetic susceptibility of the paramagnetic salt cerium magnesium nitrate, which was calibrated with two Ge resistors. The methods and whole apparatus are explained in more detail in Ref. 6.

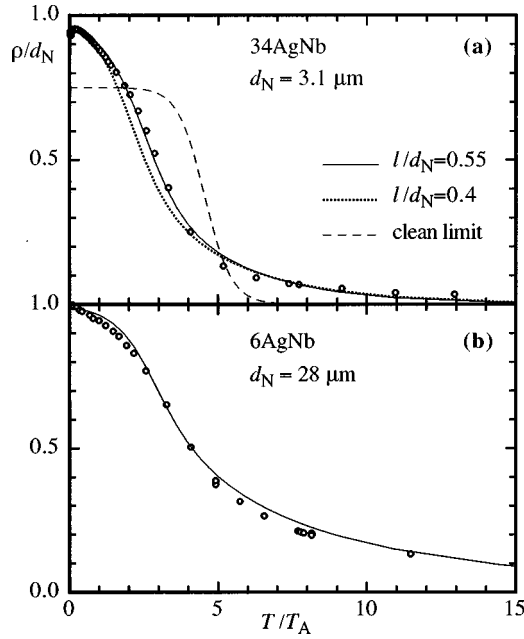


FIG. 2. Susceptibility data ( $\circ$ ) and numerical fits (solid lines) of silver-niobium samples as functions of  $T/T_A$ . (a) Fitting parameter  $l/d_N=0.55$ . The dashed line denotes the clean limit and the dotted line denotes the  $l/d_N=0.4$  curve for comparison. (b) Fitting parameter  $l/d_N=0.25$ .

#### IV. LINEAR-MAGNETIC RESPONSE

##### A. Experimental results

In the following we report on the temperature dependent ac susceptibility without external dc field, which we will address as a linear response. We show here the results for some typical samples, which represent different behaviors and sample parameters, namely annealed silver–niobium samples with a large measured mean-free path  $l_N$  as well as not annealed ones with smaller  $l_N$ . Some of them have a ratio  $d_N/r_S$  of 1, the normal layer having a rather strong curvature, while others have a ratio 0.4 and therefore an almost flat normal layer. Also shown are not annealed copper–niobium samples with ratios of 1 and 0.2 and a relatively smaller mean-free path  $l_N$  as well as two copper–tantalum samples with a ratio  $d_N/r_S=1$ .

The data of the diamagnetic screening fraction  $\rho(T)/d_N$  of the N layer are shown in Figs. 2–6 as a function of temperature in units of the Andreev temperature  $T_A$  for each sample. The rather strong development of the induced superconductivity in the normal metal (right below the transition temperature  $T_c$  of the superconductor) can be observed quite clearly.

From the susceptibility  $\chi(T)$  measured in arbitrary units the screening fraction was obtained as

$$\rho(T)/d_N = r_S/d_N \cdot [(1 + \chi_N(T)/\Delta\chi_S(T_c))^{1/2} - 1].$$

Here,

$$\chi_N(T)/\Delta\chi_S(T_c) = [\chi(T) - \chi_0]/(\chi_0 - \chi_\infty)$$

is the fraction of the temperature-dependent susceptibility of the normal metal with respect to the total diamagnetic transition of the superconductor at its critical temperature  $T_c$ .

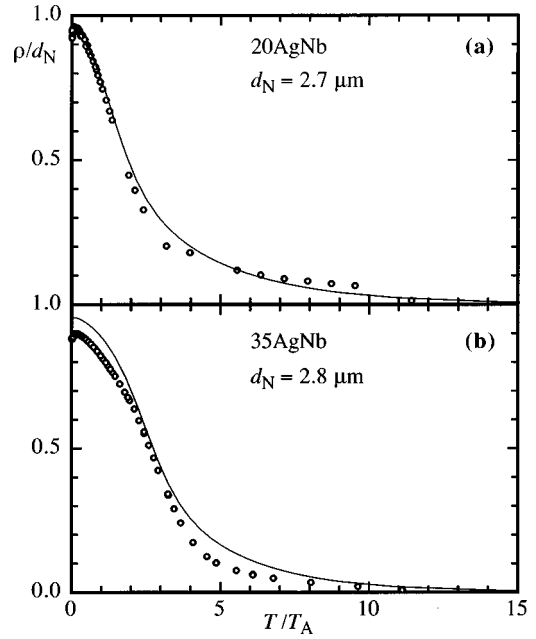


FIG. 3. Susceptibility data ( $\circ$ ) and numerical fits (solid lines) of silver-niobium samples as functions of  $T/T_A$ . Fitting parameters (a)  $l/d_N=0.25$ . and (b)  $l/d_N=0.55$ .

The exact height of both  $\Delta\chi_S$  and  $\chi_N(T)$  depends on the value of the susceptibility  $\chi_0$  right below the transition of the superconductor. Since that transition for the samples containing niobium is outside of our measuring temperature range, values at the highest temperatures were used for extrapolation of  $\chi$  to  $\chi_0$ . The susceptibility above the superconducting transition  $\chi_\infty$  was obtained from background measurements. Through the ratio of the radius of the superconductor  $r_S$  to the normal layer thickness  $d_N$  the normal layer curvature of the composite cylindrical sample is taken into account.

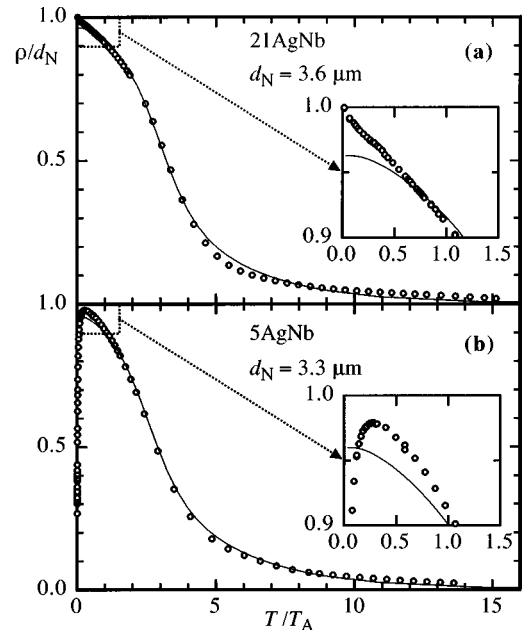


FIG. 4. Susceptibility data ( $\circ$ ) and numerical fits (solid lines) of silver-niobium samples as functions of  $T/T_A$ . Fitting parameters (a)  $l/d_N=0.75$  and (b)  $l/d_N=0.55$ . The insets are zooms in the low-temperature region for both samples.

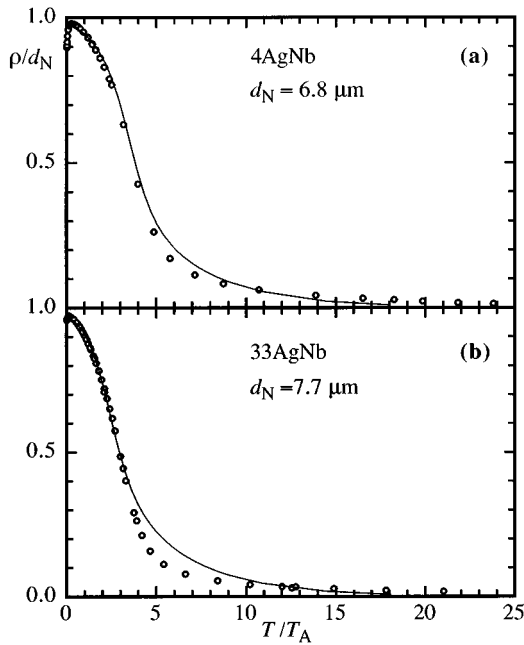


FIG. 5. Susceptibility data ( $\circ$ ) and numerical fits (solid lines) of silver-niobium samples as functions of  $T/T_A$ . (a) ratio  $d_N/r_S = 0.4$  and fitting parameter  $l/d_N = 0.75$ . (b) ratio  $d_N/r_S = 1$  and fitting parameter  $l/d_N = 0.4$ . The fit for sample 33AgNb shows a bigger deviation from the data for  $5 \leq T/T_A \leq 15$  than the fit for sample 4AgNb.

In a local picture as given by the Orsay Group on Superconductivity<sup>9</sup> the screening fraction  $\rho/d_N$  represents the screening length normalized to the normal layer thickness, with  $\rho$  the thickness of the part of the normal metal, out of which the magnetic flux is screened. For our samples that

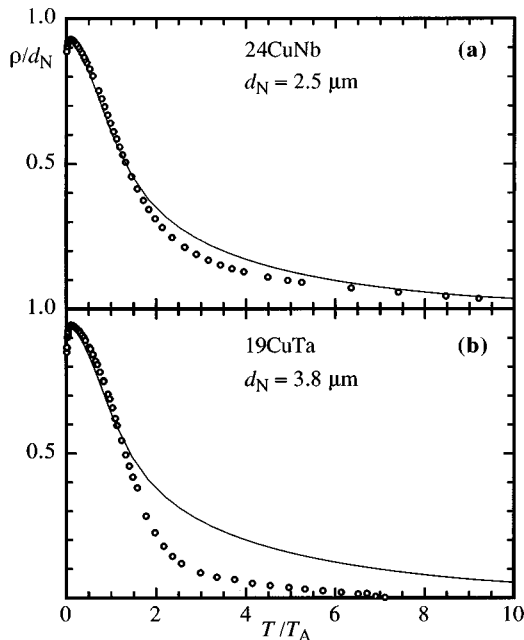


FIG. 6. Susceptibility data ( $\circ$ ) and numerical fits (solid lines) of (a) the copper-niobium sample 24CuNb, fitting parameter  $l/d_N = 0.15$  and (b) the copper-tantalum sample 19CuTa, fitting parameter  $l/d_N = 0.13$  as functions of  $T/T_A$ .

picture is not valid, since the current-field relation is nonlocal. Here, the screening fraction  $\rho/d_N$  represents the susceptibility of N, as related to a model of a one dimensional system.

### B. Discussion of the fits

The temperature dependence of the diamagnetic susceptibility of these samples is neither accounted for by the clean limit nor the dirty-limit theory. The values of the measured mean-free path  $l_N$  indicate that the samples are in an intermediate impurity regime. They are not diffusive enough for the dirty limit, because  $l_N$  is of the order of  $d_N$  for most of the samples. However, since they have a small density of scattering centers, they are not in the clean limit either. From here on we will address them as relatively clean.

We fitted the experimental data of the screening fraction with the theoretical results obtained for a one-dimensional geometry, as described in Sec. II. The fits are shown in Figs. 2–6.

The parameters  $d_N$  and  $\lambda_N$  entering the numerical calculations were obtained from measurements and well-known material constants. The only fitting parameter was then the mean-free path  $l$ , which enters into the theory through the fraction  $l/d_N$ . The fits were performed giving the experimental data at an intermediate temperature regime the most weight, where the increase of the susceptibility was the steepest. To illustrate the sensitivity of the fits to the value of the fit parameter  $l/d_N$ , in Fig. 2(a), we give in addition to the fitted curve with  $l/d_N = 0.55$ , the curve for  $l/d_N = 0.4$ .

The curves obtained from the newly developed approach with arbitrary scattering center concentration fit the experimental data well over the whole temperature range. The corresponding clean limit curve does not fit the data, as shown for comparison in Fig. 2(a).

The values of the fitting parameter  $l/d_N$  reproduce the measured values  $l_N/d_N$  rather well. It has to be emphasized that the fits are rather good, if one takes into account the difference in geometry between experiment and theory, the neglect of the boundary roughness in the theory and other imperfections inevitably present in the samples. This means, that the quasiclassical theory of the proximity effect with a finite mean-free path parameter  $l$  due to a low concentration of elastic scatterers,<sup>14</sup> is now able to explain the linear susceptibility data of our relatively clean NS specimens. A list of all the samples shown here is given in Table I. Additionally, more samples are listed together with the mean-free path  $l$ , which gave the best fit.

For the silver-niobium samples with a measured mean-free path  $l_N/d_N \approx 1$  the fitting parameter  $l/d_N$  lies between 0.4 and 0.8. The measured mean-free path of AgNb samples with  $l_N/d_N < 0.3$  is also reproduced quite well in the fits. A typical example is the silver-niobium sample 6AgNb shown in Fig. 2(b). It is surprising, that for most of the samples the agreement between the only fitting parameter  $l/d_N$  and the measured  $l_N/d_N$  is so good. However, the parameter  $l$  has to be viewed as an effective mean-free path, which contains the scattering from bulk impurities as well as from the surface. The measured mean-free path  $l_N$  is also an effective quantity, which contains surface effects, but it is yielded from transport measurements along the axis of the wires. There the

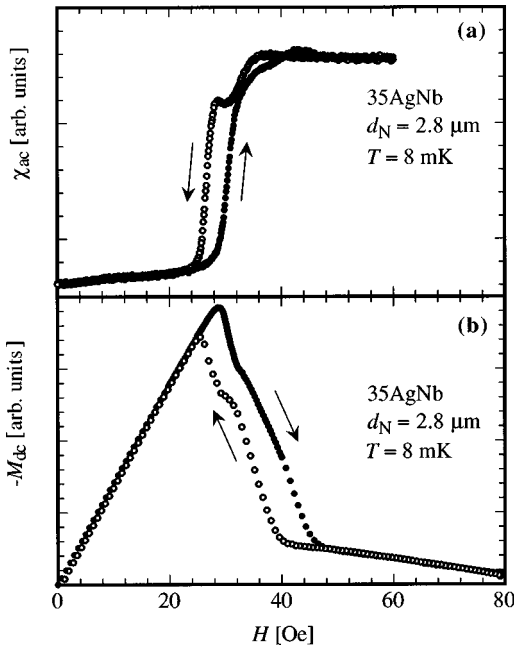


FIG. 7. (a) Nonlinear ac susceptibility as a function of magnetic field  $H$ . (b) Isothermal dc magnetization curve.

surface is expected to influence the mean-free path in a different way, because the geometry of the relevant trajectories differs from the ones in the proximity case.

In the following, we describe in detail as an example, the quality of the fits for the silver-niobium samples. These samples show the best agreement between the measured magnetic susceptibility and the theory. The results for some of them are illustrated in Figs. 2–5. In general, the fits given by the solid lines, describe the experimental data well. In particular, for some of these samples we observe considerable deviations at the lowest temperatures ( $T < T_A$ ) where the theoretical curves saturate as shown in Fig. 1. Moreover, the deviations are different for nominally similar samples. For example, sample 21AgNb [inset of Fig. 4(a)] shows higher  $\chi_N$  values than the theory, while sample 5AgNb [inset of Fig. 4(b)] shows also higher values and in addition a strong reentrance of  $\chi_N$  at the lowest temperatures. These apparently contradictory results are possibly related to the surface quality. At this point, it is not clear how a nonideal reflecting surface affects the susceptibility, neither experimentally nor theoretically. In addition, silver–niobium samples with roughly the same  $d_N$  and  $l$  show strongly different sizes of reentrance. The origin of these different behaviors will be subject of further investigations.

The fit shown in Fig. 3(b) is rather poor, which could be explained by the geometrical nonuniformity of sample 35AgNb, showing effectively two different values of  $d_N$  (see also Fig. 7 and Sec. V).

For  $T \geq T_A$  the fits of the AgNb specimens are quite good. For all the specimens, in a medium temperature regime, the theoretical susceptibility is a bit too high with respect to the measured one. This behavior can be observed more clearly for the samples in Fig. 5. Their normal layer thickness with values of  $d_N = 6.8 \mu\text{m}$  and  $d_N = 7.7 \mu\text{m}$  as well as the measured mean-free path with values of  $l_N = 5.1 \mu\text{m}$  and  $l_N = 5.3 \mu\text{m}$  are approximately the same. The only difference

between the two samples is the ratio  $d_N/r_S$ , which for sample 4AgNb is 0.4 and for sample 33AgNb is 1. As expected, sample 33AgNb with the bigger ratio and therefore with the stronger curvature of the normal layer shows a more pronounced deviation in the medium temperature regime.

The susceptibility of the copper–niobium samples is also described by the theory rather well. A typical example is shown in Fig. 6(a). For similar samples the fitted values of  $l/d_N$  are very small, lying between 0.02 and 0.2 (see Table I). In contrast to the silver-niobium samples with very small mean-free paths, for the copper-niobium samples the value of  $l/d_N$  is only about half of the measured value of  $l_N/d_N$ . The CuNb samples show similar deviations as the silver-niobium samples in the different temperature regimes.

The poorest agreement between experiment and theory is met for the copper-tantalum samples. An example is shown in Fig. 6(b). As the temperature is increased the theoretical curve starts to show much more screening than the experiment. The deviation starts to show up, where the induced diamagnetism is about half reduced. At higher temperatures the difference between the two curves is even larger. Moreover, the fitting parameter  $l/d_N$  is much smaller than the measured  $l_N/d_N$ . Interestingly, the two copper-tantalum samples investigated (14CuTa and 19CuTa) show the same strong disagreement between  $l$  and  $l_N$ . This suggests that some other effects should be considered for the combination of these two materials.

## V. NONLINEAR-RESPONSE MEASUREMENTS

We have investigated for our NS proximity samples the nonlinear-magnetic response. With the experimental setup described in Ref. 6 ac-susceptibility measurements at constant temperature as a function of dc magnetic field were performed, as well as isothermal dc-magnetization curves.

Figure 7 shows the nonlinear susceptibility and the dc-magnetization curve for one of the cleanest silver-niobium samples at  $T = 8 \text{ mK}$ . The magnetic breakdown of the induced superconductivity occurs at a magnetic field  $H_b \approx 25 \text{ Oe}$ . Below the breakdown field the magnetic flux is totally screened, and above, it enters into the normal metal layer. The first-order phase transition shows the features of superheating at increasing field and of supercooling at decreasing field. That kind of hysteretic behavior was already observed and discussed in the dirty limit by the Orsay Group on Superconductivity.<sup>9</sup> This particular sample shows a small second transition, which is due to the niobium core not being perfectly centered resulting in effectively two slightly different normal layer thicknesses. We notice that the magnetic breakdown transition is not infinitely sharp. In our samples consisting of several hundred wires the superheated and supercooled transitions are triggered for each individual wire at slightly different fields, leading to a statistical broadening of the breakdown jumps.

From the isothermal  $\chi_N(H)$  curves we have determined the supercooled and superheated fields as the fields at the middle of each transition. In this way, values of  $H_{sc}$  and  $H_{sh}$  at different temperatures were obtained.

For our relatively clean silver-niobium samples at temperatures higher than the Andreev temperature  $T_A \propto v_F/d_N$ ,

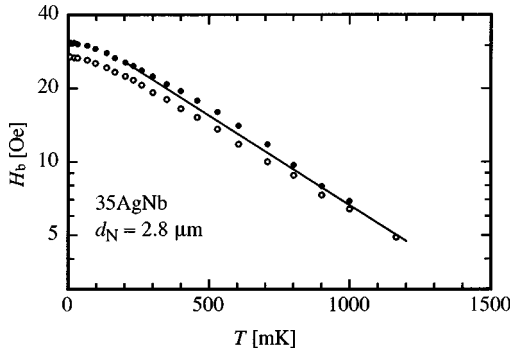


FIG. 8. Measured supercooled ( $\circ$ ) and superheated ( $\bullet$ ) breakdown fields of the annealed silver-niobium sample 35AgNb with  $d_N = 2.8 \mu\text{m}$ . The line represents the clean-limit theory of a one dimensional NS slab with a correction factor 0.3.

the experimental breakdown fields follow the exponential dependence  $H_b(T) \propto 1/d_N \exp[-d_N/\xi_N(T)]$ . The experimentally found values of  $\xi_N(T)$  reproduce the theoretical clean limit coherence length in silver  $\xi_T = \hbar v_F / 2\pi k_B T = 1.69 \mu\text{m}/T(\text{K})$  within a few percent. For sample 35AgNb, this is illustrated in Fig. 8, where the experimental supercooled and superheated fields as a function of temperature are given. The theoretical clean limit breakdown field of a one-dimensional NS slab for  $T \gg T_A$  is shown as a solid line. A factor of about 0.3 has been used to shift down the theoretical curve to fit the experiment. We notice that the temperature dependence of the breakdown field agrees qualitatively with the clean-limit result,<sup>15</sup> obtained for a one-dimensional NS slab assuming ideal boundary conditions, e.g., ideal normal electron transmission at the NS interface. Quantitatively, the measured breakdown fields are a factor of about 0.3 smaller than the high-temperature theoretical curve (see Fig. 8).

This discrepancy could have different reasons. First, in our samples normal reflections at the NS interface may happen due to the mismatch of the Fermi velocities between N and S and impurities or interdiffusion. Second, our samples have a cylindrical geometry, which weakens the proximity effect with respect to the one-dimensional flat geometry considered in the theory. And third, the influence of impurity scattering on the degree of nonlocality is expected to lead to quantitative deviations of the data from the clean-limit theory.

In Fig. 9 the saturated superheated fields of the silver-niobium samples are plotted versus the inverse normal layer thickness  $1/d_N$ . The samples with  $l_N/d_N \approx 1$  and  $d_N/r_S = 0.4$  show a clear  $1/d_N$  dependence of the breakdown field, in agreement with the clean-limit theory.<sup>15</sup> On the other hand, the absolute values of the saturated breakdown fields are about a factor of 1.7 lower than in the clean-limit theory. All the other samples with a ratio  $d_N/r_S = 1$  or with a lower mean-free path  $l_N$  show smaller breakdown fields. As already briefly mentioned, for the copper samples the influence of a reduced normal electron transmission coefficient at the NS boundary could be especially strong leading to a further suppression of the breakdown field.

The silver-niobium samples with  $l_N/d_N < 0.3$  and the copper samples show a temperature dependence  $H_b(T) \propto \exp[-d_N/\xi_N(T)]$ , but for these samples experimentally we found  $\xi_N(T) = p \cdot \xi_T$ , where the prefactor  $p$  is about 0.3 to 0.6.<sup>6</sup> The

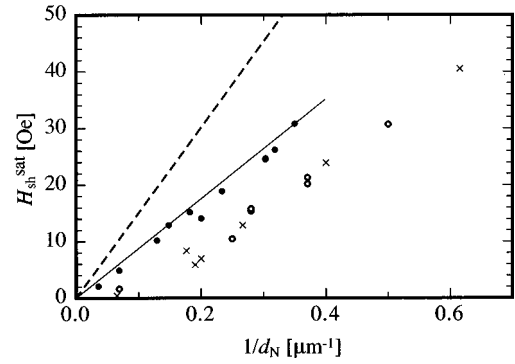


FIG. 9.  $1/d_N$  dependence of the saturated superheated field for silver-niobium samples with  $l_N/d_N \approx 1$  ( $\bullet$ ). Slope of solid line:  $88 \text{ Oe } \mu\text{m}$ . The dashed line represents the clean-limit result, with slope:  $150 \text{ Oe } \mu\text{m}$ . Also shown are less clean silver-niobium samples with  $l_N/d_N < 0.3$ , ( $\circ$ ) and copper samples ( $\times$ ), which we do not consider in the fit.

breakdown field data of these samples cannot be explained by the clean-limit theory, since impurity scattering is too strong.

Although the temperature dependence of the breakdown field is the same as given by the clean limit, the full form of the magnetization curves  $M(H)$  of the cleanest AgNb samples cannot be explained within this limit. The slope below the transition does not correspond to the  $-3/4$  of a perfect diamagnet and additionally, the observed finite screening tail above the transition [see Fig. 7(b)] does not appear in the clean limit. Clearly, the correct description of the magnetization data with the quasiclassical theory should take into account the finite mean-free path  $l$ . [Note the similarity between the magnetization curve in Fig. 7(b) and the theoretical dirty limit magnetization curves from Ref. 13, which have the right slope as well as a finite screening tail, but give the wrong field scale.] Theoretical work on  $M(H)$  for arbitrary low impurity concentrations is still needed.

## VI. CONCLUSIONS

We investigated the magnetic response of NS proximity layers of high purity, and compared the experimental data with the quasiclassical theory. A large number of samples containing combinations of Ag and Cu with Nb and Ta has been fabricated and measured. All the samples show an induced diamagnetic susceptibility close to  $-1/4\pi$  for temperatures  $T \lesssim T_A$ . The mean-free path  $l_N$  obtained from resistivity measurements varied between  $0.1d_N$  and  $1.3d_N$ . With the results of Belzig, *et al.* which are based on the quasiclassical theory including arbitrary impurity concentrations, we were able to reproduce the experimental data quite well, using a mean-free path  $l$  as the only fitting parameter. The mean-free path  $l$  obtained in this way agrees within a factor of 2 with the mean-free path  $l_N$  determined by resistivity measurements. This good agreement shows that the linear-diamagnetic response of a proximity system with arbitrary impurity concentration is very well described with the present nonlocal approach. However, the very low-temperature behavior, where some deviations occur and in

addition an unexplained reentrance of the susceptibility appears, is still not understood.

Magnetization measurements show a first-order transition at a breakdown field from a state with almost complete flux expulsion to a state with weak screening. The temperature dependence of the breakdown field for the cleanest specimens is in accordance with the clean limit. Nevertheless, the complete magnetization curve, in agreement with the linear susceptibility data, reflects the finite mean-free path of the samples.

With this work we show that the proximity theory, based on the quasiclassical approximation including the full nonlocal current response, can successfully describe the magnetic response of very clean samples. The experimentally unavoid-

able low level of impurities can be accounted for with a mean-free path  $l$ , which is of the order of the sample size.

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<sup>1</sup>R. Holm and W. Meissner, *Z. Phys.* **74**, 715 (1932).

<sup>2</sup>G. Deutscher and P. G. de Gennes, in *Superconductivity*, edited by R. D. Parks (Dekker, New York, 1969), Vol. 2, p. 1005.

<sup>3</sup>A recent review of the proximity effect in mesoscopic diffusive conductors is found in *Mesoscopic Electron Transport*, Vol. 345 of *NATO Advanced Studies Institute, Series E: Applied Sciences*, edited by L. L. Sohn, L. P. Kouwenhoven, and G. Schön (Kluwer Academic Publishers, Dordrecht, 1997).

<sup>4</sup>A. C. Mota, D. Marek, and J. C. Weber, *Helv. Phys. Acta* **55**, 647 (1982); J. C. Weber, A. C. Mota, and D. Marek, *J. Low Temp. Phys.* **66**, 41 (1987).

<sup>5</sup>Th. Bergmann, K. H. Kuhl, B. Schröder, M. Jutzler, and F. Po-bell, *J. Low Temp. Phys.* **66**, 209 (1987).

<sup>6</sup>A. C. Mota, P. Visani, and A. Pollini, *J. Low Temp. Phys.* **76**, 465 (1989).

<sup>7</sup>P. Visani, A. C. Mota, and A. Pollini, *Phys. Rev. Lett.* **65**, 1514 (1990); A. C. Mota, P. Visani, A. Pollini, and K. Aupke, *Physica B* **197**, 95 (1994).

<sup>8</sup>L. N. Cooper, *Phys. Rev. Lett.* **6**, 689 (1961).

<sup>9</sup>Orsay Group on Superconductivity, in *Quantum Fluids*, edited by D. Brewer (North-Holland, Amsterdam, 1966).

<sup>10</sup>Y. Oda and H. Nagano, *Solid State Commun.* **35**, 631 (1980).

<sup>11</sup>A. D. Zaikin, *Solid State Commun.* **41**, 533 (1982).

<sup>12</sup>A. F. Andreev, *Zh. Eksp. Teor. Fiz.* **49**, 655 (1965) [*Sov. Phys. JETP* **22**, 455 (1966)].

<sup>13</sup>W. Belzig, C. Bruder, and G. Schön, *Phys. Rev. B* **53**, 5727 (1996).

<sup>14</sup>W. Belzig, C. Bruder, and A. L. Fauchère, *Phys. Rev. B* **58**, 14 531 (1998).

<sup>15</sup>A. L. Fauchère and G. Blatter, *Phys. Rev. B* **56**, 14 102 (1997).

<sup>16</sup>C. Bruder and Y. Imry, *Phys. Rev. Lett.* **80**, 5782 (1998).

<sup>17</sup>G. Eilenberger, *Z. Phys.* **214**, 195 (1968).

<sup>18</sup>A. I. Larkin and Yu. N. Ovchinnikov, *Zh. Eksp. Teor. Fiz.* **55**, 2262 (1968) [*Sov. Phys. JETP* **28**, 1200 (1969)].

<sup>19</sup>For a recent review of the quasiclassical formalism see *Quasi-classical Methods in Superconductivity and Superfluidity*, edited by D. Rainer and J. A. Sauls (Springer, Heidelberg, 1998).

<sup>20</sup>P. G. de Gennes and D. Saint-James, *Phys. Lett.* **4**, 151 (1963).

<sup>21</sup>K. D. Usadel, *Phys. Rev. Lett.* **25**, 507 (1970).

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