Pair-breaking mechanisms in superconductor—normal-metal—superconductor junctions

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The critical current density J_c has been measured for superconductor—normal-metal—superconductor (S-N-S) junctions over a wide range of temperature and composition in order to determine the depairing effects of magnetic impurities. Junctions, which are in a sandwich geometry with the N layer typically 600 nm thick, show well-defined diffraction patterns indicating that the junctions are of high quality. Below 4.2 K, the temperature dependence of J_c is found to follow a modified bridge theory based on the work of Makeev *et al.* (Fiz. Nizk. Temp. **6**, 429 (1980) [Sov. J. Low Temp. Phys. **6**, 203 (1980)]). In this range, the coherence length and order parameter in the superconductor are essentially independent of temperature, and so it is reasonable that the bridge and sandwich geometry results are similar. As the temperature approaches the transition temperature (T_{cS}) of the superconductor, J_c was found to be proportional to $(1-T/T_{cS})^2$ as predicted by de Gennes.

I. INTRODUCTION

Superconductor—normal-metal—superconductor (S-N-S) junctions have been widely used to study the transport properties of electrons in proximity effect systems. From the temperature dependence of the critical current density, for example, one can determine the pair potential,[1–3] the depairing parameters of magnetic impurities,[4] and various quantum effects[5] related to the N layers. Clarke[3] extensively studied the zero-field temperature dependence of the critical current densities J_c for S-N-S junctions, where the N layer contained nonmagnetic impurities and was in the dirty limit. Hsiang and Finnemore[6] then extended those studies to S-N-S junctions where the N layer was in the clean limit. In both studies, J_c was found to vary with the temperature T according to

$$J_c = J_0(1-T/T_{cS})^2 \exp(-K_N d_N) , \qquad (1)$$

where J_0 is a constant, T_{cS} is the transition temperature of the S layer, K_N^{-1} is the pair penetration depth in the N layer, and d_N is the junction thickness. At low temperature, they also found that K_N was proportional to $T^{1/2}$ in the dirty limit[3,4] and K_N was proportional to T in the clean limit.[6]

For S-N-S junctions in which the N layer contains magnetic impurities, relatively little work has been done on the temperature dependence of J_c. Paterson[4] studied the temperature dependence of J_c for S-N-S junctions with CuNi and CuMn barriers over a relatively small reduced temperature range from $T/T_{cS}=0.15$ to 0.58. The measured J_c, however, was only approximately represented by the expression derived by Paterson,

$$J_c = B \exp\{-\gamma[(T+\alpha)/T_c]^{1/2}\} , \qquad (2)$$

where B, γ, and α are constants, and T_c is the transition temperature of the junction. For CuMn, in fact, the data fitted

$$J_c = B \exp\{-\gamma[(T+\alpha)/T_c]^{3/2}\} .$$

It was clear that further work was needed over a wider temperature range to establish the correct theoretical framework.

The purpose of this work is to study the transport properties of electrons in Pb-$Ag_{1-x-y}Mn_xAl_y$-Pb junctions both in concentrated and dilute limits in order to determine the way in which J_c depends on the spin-scattering mean free path l_{sp} and the transport mean free path l_N in the N layer. At low temperature where the coherence distance in the superconductor, ξ_S, is independent of temperature, the value of the pair potential at the superconductor—normal-metal (S-N) interface should be independent of T and the theory of Makeev *et al.*[1] should apply for both the bridge geometry and the sandwich geometry. The goal is to test this idea and to determine the range of validity of the de Gennes theory[2] near T_c.

II. EXPERIMENTAL PROCEDURES

The $Ag_{1-x}Mn_x$ alloys were made by arc-melting Ag (99.999% pure) with Mn (99.995% pure) and then annealing in an argon atmosphere. The dilute $Ag_{1-x-y}Mn_nAl_y$ alloys were prepared by arc-melting a small amount of the concentrated $Ag_{1-x}Mn_x$ alloy with pure Ag and Al and then annealing in an argon atmosphere. The samples were then rolled between two molybdenum foils into a sheet $\sim$0.2 mm thick. The sheet was then cut into small pellets which were then used to prepare the S-N-S junctions.

The cross-type S-N-S junctions[5] were prepared by depositing thin films onto a glass substrate (25.4×12.7 mm^2) in a high-vacuum (1×10^{-8} Torr) evaporator system. The bottom Pb strip was first evaporated from an electrically heated molybdenum boat. The Pb film had a thickness of about 700 nm and a mean free path (l_S) of $\sim$360 nm determined from the resistivity using the relation[7] $(\rho l_S)^{-1}=9.4\times10^{10}\ \Omega^{-1}\mathrm{cm}^{-2}$. The normal barrier, which was effectively square with a junction width of $\sim 7\times10^{-3}$ cm, was prepared by subsequently dropping the tiny alloy pellets into a hot molybdenum boat and completely evaporating each pellet, giving a thickness increase per pellet of about 5 nm. The thickness of the N layers varied from ~ 200 to 600 nm. During evaporation the substrates were cooled to a temperature of about $=30\,^\circ\mathrm{C}$ by pumping ethyl alcohol out of the cold trap which was attached to the substrates. Pumped alcohol rather than liquid nitrogen was used to cool the substrates in preparing the samples because the N layer tends to crack when films deposited at 78 K are warmed to room temperature. A very slow warming rate of a few degrees per hour is needed to prevent cracking. For each evaporated S-N-S junction, a single alloy strip was simultaneously deposited on another substrate and was used to measure the resistivity and thickness of the N layer. The current through the junction was symmetric. A sensitive rf-SQUID (superconducting quantum-interference device[8,9]) voltmeter was used to obtain an accurate critical current. The current-voltage characteristic was recorded on an x-y recorder by feeding the output of a low-noise current source and the output of the SQUID into the x and y axes of the recorder, respectively. The voltage resolution was $\sim 1\times 10^{-12}$ V. A superconducting magnet was used to apply a magnetic field to the sample. The ambient field was shielded by two layers of μ-metal outside the nitrogen Dewar and a superconducting cylinder outside the vacuum can. The magnetic field was applied parallel to one of the Pb strips and parallel to the plane of the normal layer. In the study of the magnetic field dependence of the critical current (I_c-versus-H curves), the field was applied to the sample while the temperature of the sample was above the transition temperature T_c of the junction and the junction was cooled in the field through T_c. The temperature was measured with a calibrated germanium thermometer.

III. RESULTS AND DISCUSSIONS

A. Properties of the normal layers

The data characterizing the normal (N) layers for different samples are summarized in Table I. The resistivities at 300 and 4.2 K are those calculated from the measured resistances of the adjacent metal strips. These resistivities for current parallel to the film may not be the correct ones to use to calculate the electronic mean free path l_N for electrons moving perpendicular to the film, but they are a first indication. The l_N value at 4.2 K was calculated from $(\rho l_N)^{-1}=8.6\times 10^{10}\ \Omega^{-1}\,\mathrm{cm}^{-2}$,[6] where ρ is the resistivity. For a first look at the data, we assume this is also the mean free path that controls the pair diffusion into the N layer. The l_N value is somewhat smaller than the coherence lengths at 7 K, which indicates all the N layers are in the dirty limit.

B. Current-voltage characteristics

Current-voltage (I-V) characteristics[10,11] for these junctions follow a resistively-shunted-junction (RSJ) model rather well and are similar to results reported earlier.[6] There are no voltage steps or kinks in the curves. Possibly the most important feature of the I-V curves is that they are totally reversible and show no signs of flux pinning or flux trapping as long as the field is parallel to the junction. Flux moves through the normal layers with great ease even though there are substantial supercurrents.

C. Magnetic field dependence of I_c

To test the quality of the junction, the magnetic field dependence of the maximum supercurrent was taken for each sample. A sample which has both a uniform junction and uniform current distribution should show a good Fraunhofer diffraction pattern for the temperature region where the Josephson penetration depth[12,13] λ_J is greater than $\frac{1}{4}$ of the junction width. Figure 1 shows an I_c-versus-H curve for a sample at 4.52 K for which $W/\lambda_J \simeq 0.86$. The solid curve is the Fraunhofer diffraction pattern, using

$$I_c \propto \left| \frac{\sin(\pi\phi/\phi_0)}{(\pi\phi/\phi_0)} \right| ,$$

which is normalized to the zero-field I_c maximum and the third I_c minimum at 1.14 G. Here $\phi = HW(d_N+2\lambda_S)$ is the total flux threading the junction,

TABLE I. Properties of normal (N) layers. The electronic mean free path l_N was calculated from $(l_N\rho)^{-1}=8.6\times 10^{10}\ \Omega^{-1}\,\mathrm{cm}^{-2}$, where ρ is the resistivity. The last two columns are the coherence lengths from the dirty- and clean-limit formulas. The Fermi velocity of Ag used in calculation is $v_{FN}=1.38\times 10^{6}$ m/sec.

Sample no.	Alloy	Mn content (at. %)	Al content (at. %)	Thickness (nm)	$\dfrac{\rho(300\ \mathrm{K})}{\rho(4.2\ \mathrm{K})}$	l_N (4.2 K) (nm)	$\left[\dfrac{h v_{FN} l_N}{6\pi k_B T}\right]^{1/2}$ at 7 K (nm)	$\dfrac{h v_{FN}}{2\pi k_B T}$ at 7 K (nm)
1	Ag-Mn	2	0	220	1.56	37.0	54.4	240
3	Ag-Mn	1.5	0	320	1.37	31.9	50.5	240
4	Ag-Mn	1.5	0	345	1.89	41.5	57.6	240
5	Ag-Mn	1.5	0	295	1.73	34.7	52.7	240
6	Ag-Mn	1.5	0	280	1.82	38.8	55.7	240
7	Ag-Mn	0.8	0	450	2.09	68.4	73.9	240
8	Ag-Mn	0.8	0	500	2.16	67.6	73.5	240
A	Ag-Mn-Al	0.073	6.4	250	1.31	16.6	36.4	240
B	Ag-Mn-Al	0.073	6.4	430	1.31	15.6	35.3	240

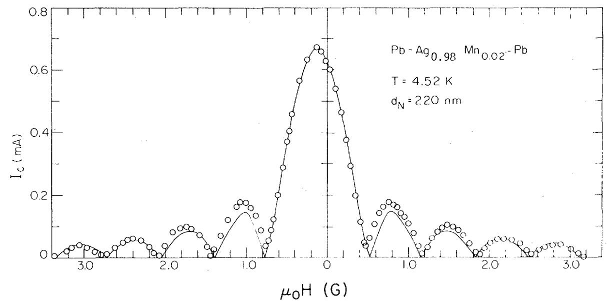

FIG. 1. Pb-$Ag_{0.98}Mn_{0.02}$-Pb; $W/\lambda_J = 0.86$.

where ϕ_0 is the flux quantum, W is the barrier width, and λ_S is the penetration depth in the superconductor. The data show a nice fit to the theoretical prediction except that the first two maxima rise slightly above the theoretical values. If the flux quantum is taken to be $2\times10^{-7}\,\mathrm{G\,cm^2}$ and $\Delta\phi = \Delta H W(d_N + 2\lambda_S)$, then the measured values of ΔH, W, and d_N give $\lambda_S = 120$ nm at 4.7 K. This is slightly larger than bulk Pb, as would be expected near an S-N interface where N is doped with magnetic impurities. The central maximum of the I_c-versus-H curve has been shifted from $H = 0$ to a small negative value due to a partially asymmetric current feed caused by the induced current in the superconducting input Pb wires. The I_c-versus-H curve is symmetric with respect to the central maximum, which indicates the homogeneity of the sample. As λ_J decreases relative to W, the minima are lifted and the sample shows a partial Meissner effect at low field.

D. Zero-field temperature dependence of J_c

Figure 2 shows the temperature dependence of the critical current density J_c for several junctions. The J_c value increases monotonically as the temperature decreases and reaches saturation at low temperature. This saturation behavior is similar to the theoretical prediction for superconductor-insulator-superconductor (S-I-S) junctions[14] and S-N-S bridges.[15]

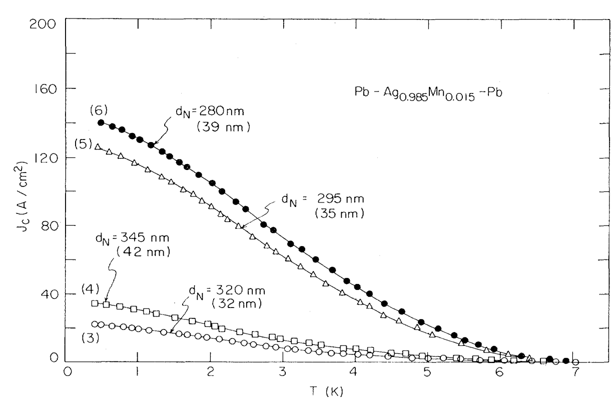

FIG. 2. J_c vs T for Pb-$Ag_{1-x}Mn_x$-Pb junctions. The numbers in parentheses refer to its electronic mean free path. D_N and l_N dependence of J_c for Pb-$Ag_{0.985}Mn_{0.015}$-Pb junctions.

Although the theory of Makeev *et al.*[1] does not exactly apply for this experiment, it is possible to modify the theory so that the conditions are rather close. Makeev *et al.* calculated the Josephson supercurrent at any arbitrary temperature for S-N-S junctions with paramagnetic impurities in both superconducting and normal metals. The calculations were performed with the aid of the Usadel equations[16] and the J_c expression derived as follows:

$$J_c = \frac{4\pi\sigma_N k_B T}{e}\sum_{\omega}\frac{2\tilde{\omega}}{D_N}[1 - y(d_N/2)]\exp\left[-d_N\frac{2\tilde{\omega}}{D_N}\right], \tag{3}$$

where

$$\tilde{\omega} = |\omega| + 1/\tau_s, \tag{4}$$

$\omega = \pi k_B T(2n+1)/h$ is the Matsubara frequency, $D_N = \frac{1}{3}v_{FN}l_N$, σ_N is the electrical conductivity, k_B is Boltzmann's constant, e is the electron charge, τ_s is the spin-flip—scattering time, and $y(d_N/2)$ is a complicated function of Δ_s, ω, and τ_s.[1] In the notation of Makeev *et al.*, the function $y(Z)$ is actually the Usadel Green's function, $G(\omega,\tau_s,Z)$ (Ref. 16) and it should be continuous at the boundary, i.e., $y(d_N/2)$ is equal to its equilibrium value in S, $G_S(\omega,\tau_s)$.

These, of course, are not the exact conditions of our experiment and a modification is needed in order to apply Eq. (3) to our case. For this experiment only the N layer contains magnetic impurities, which would imply that $\tau_s = \infty$ in S. In the absence of magnetic impurities in S, $y(d_N/2)$ is equal to the equilibrium value in S, i.e.,

$$y(d_N/2) = \frac{\hbar\omega}{(\Delta^2 + \hbar^2\omega^2)^{1/2}}, \tag{5}$$

where $\Delta_S(T)$ is the order parameter in S. One then obtains the J_c expression,

$$J_c = \frac{4\pi\sigma_N k_B T}{e}\sum_{\omega}\frac{2\tilde{\omega}}{D_N}\left[1 - \frac{\hbar\omega}{(\Delta_s^2 + \hbar^2\omega^2)^{1/2}}\right] \times\exp\left[-d_N\frac{2\tilde{\omega}}{D_N}\right], \tag{6}$$

where

$$2\tilde{\omega}/D_N = \xi_c^{-1}[(T/T_c)(2n+1) + \hbar/(\pi k_B/T_c\tau_s)]^{1/2}, \tag{7}$$

$$\xi_c = \left[\frac{\hbar_{FN} l_N}{6\pi k_B T_c}\right]^{1/2}, \tag{8}$$

and T_c is the transition temperature of the superconductor.

Using this modified-bridge theory we have fitted the low-temperature J_c data to Eq. (6). The reduced length d_N/ξ_C and τ_s in that equation were chosen for a best fit to the shape of J_c at low temperature. This is indicated by the solid curve in Fig. 3. The derived d_N/ξ_C and τ_s from the fit to the data at low temperature along with the values calculated from thickness and l_N data, labeled $(d_N/\xi_C)_{\rm calc}$, are shown in Table II. One can see a good fit to the data at low temperature which shows that the sandwich junctions have the same temperature depen-

TABLE II. Parameters for the S-N-S junctions reported here. The d_N/ξ_C and τ_S are from fits to the low-temperature data, along with the calculated $(d_N/\xi_C)_{calc}$ values. Also shown are the τ_S's obtained from fits to the data near T_c using Kogan theory and the δ values, where $\delta = h/\pi k_B \tau_S$.

Sample no.	d_N/ξ_C	$(d_N/\xi_C)_{calc}$	τ_S Makeev *et al.* (10^{-13} sec)	τ_S Kogan (10^{-13} sec)	$\delta = h/\pi k_B \tau_S$ Kogan (K)
1	12.0	4.07	3.03	2.45	9.9
3	13.7	6.39	3.13	3.41	7.1
4	13.7	6.04	3.39	3.24	7.5
5	11.8	5.67	2.95	2.65	9.2
6	11.5	5.11	2.87	2.50	9.7
7	12.0	6.13	3.01	3.22	7.55
8	14.7	6.84	4.20	4.51	5.4
B	22.0	12.35	13.1		

dence for J_c as the bridge junctions. The value of $(d_N/\xi_C)_{calc}$ differs from d_N/ξ_C derived from I_c by a factor of 2 rather consistently. At low temperature, the coherence length ξ_S is essentially independent of temperatures so that the order parameter at the boundary, Δ_S, and the order parameters in bulk, Δ_S (bulk), are both independent of temperature. This is just the situation required by the theory.[1] Δ_S is simply somewhat smaller than Δ_S(bulk). Hence, we interpret the good fit to the data to indicate that the modulus of the order parameter at the boundary is indeed proportional to its bulk value in the superconductor as required by the Makeev theory.[1] Note that the data near T_c are quite different from the bridge theory. For a bridge structure the theory predicts a linear dependence in J_c versus T for T near T_c, whereas the S-N-S—sandwich data show a linear dependence in $J_c^{1/2}$ versus T as expected from de Gennes.[2] The full implications of the theory of Makeev *et al.* have been presented elsewhere.[17]

Figure 4 shows the temperature dependence of $J_c^{1/2}$ for several samples of different Mn concentrations over the full temperature range. The dominant feature of this plot is the linear dependence of $J_c^{1/2}$ over a wide temperature range for the 1.5 and 2.0-at. % Mn samples. Theoretically, we expect this linear dependence of $J_c^{1/2}$ to occur for T close to T_c as predicted by de Gennes.[2] The data show that the high-Mn-concentration samples follow this behavior to a reduced temperature (T/T_c) well below 0.5.

Near T_c all of the samples follow de Gennes as illustrated by Fig. 5, which shows the $J_c^{1/2}$-versus-T plot for several samples. In order to extract τ_S from the data near T_c, a sandwich theory based on work by Kogan[18] was used. Kogan derived the temperature dependence of J_c for S-N-S junctions where the N layer contains magnetic impurities and the S layers are in the clean limit. He obtained the following expression:

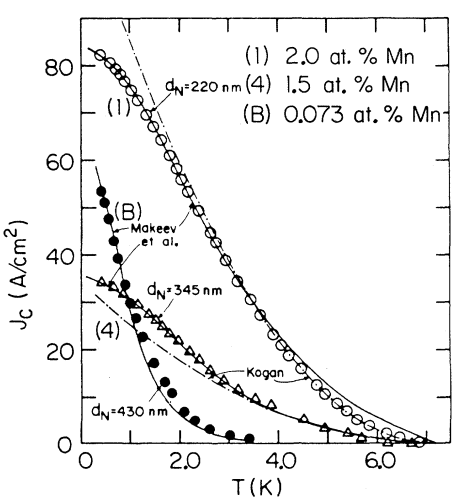

FIG. 3. Temperature dependence of J_c for Pb-$Ag_{0.98}Mn_{0.02}$-Pb, Pb-$Ag_{0.985}Mn_{0.015}$-Pb, and Pb-$Ag_{1-x-y}Mn_xAl_y$-Pb junctions, where $x = 0.000\,73$ and $y = 0.064$. The solid lines are from fits to the low-temperature data using the bridge theory. The dashed lines are from fits to the data near T_c using Kogan theory.

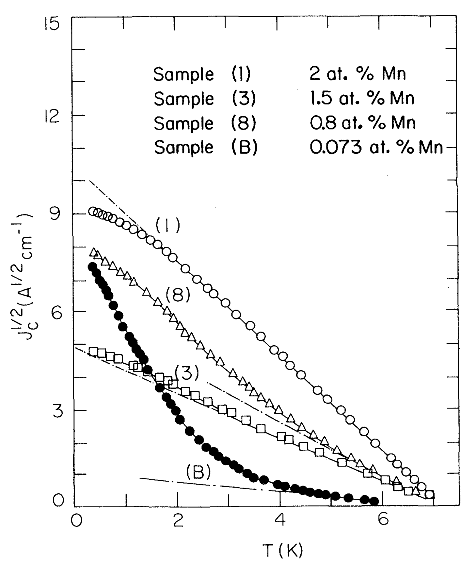

FIG. 4. $J_c^{1/2}$ as a function of temperature for several samples showing the variation of $J_c^{1/2}$ with concentration. $d_N = 2200$ Å for sample 1, 5000 Å for sample 8, 320 nm for sample 3, and 430 nm for sample B.

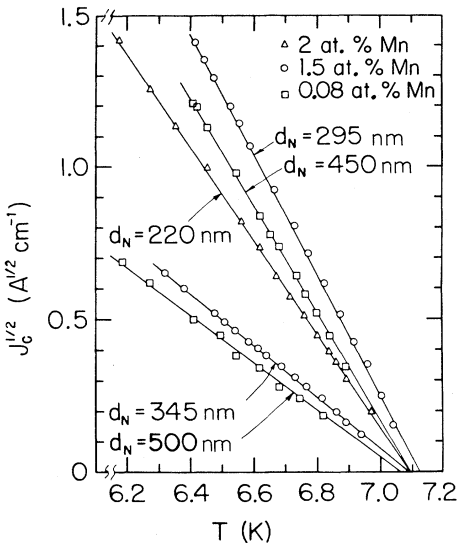

FIG. 5. $J_c^{1/2}$ vs T for several samples.

$$J_c=\frac{16(12\pi\sigma_N k_B)}{[7\zeta(3)]^2 e\tau_N^2 v_{FS} K_N T_{cS}}(T_{cS}-T)^2\exp(-K_N d_N)\,, \qquad (9)$$

where K_N is given by

$$K_N=\xi_c^{-1}[T/T_c+h/(\pi k_B T_c\tau_S)]^{1/2} \qquad (10)$$

and

$$\xi_c=(hv_{FN}l_N/6\pi k_B T_c)^{1/2}\,, \qquad (11)$$

$\zeta(3)=1.202$ is a value of the ζ function, T_{cS} is the transition temperature of the superconductor, τ_N+l_N/v_{FN}.

Because J_c varies with temperature according to

$$J_c\propto(1/K_N)(T_{cS}-T)^2\exp(-K_N d_N)\,, \qquad (12)$$

and K_N changes very slowly with temperature as T approaches T_{cS}, we should expect a linear dependence in $J_c^{1/2}$ versus T. There is only one unknown in Eq. (9), namely τ_s; therefore, if we fit the data near T_c to Eq. (9), τ_S can be obtained experimentally. The τ_S values obtained from the data are shown in Table II. Here we have used the derived d_N/ξ_S obtained from fits to the modified-bridge theory to derive the τ_S's. One can see that the τ_S derived from data near T_c is consistent with that from the low-temperature data.

In the planning of this experiment it was expected that rather large changes in τ_S would occur for the 0.8-at. % Mn sample near 4 K where there is supposed to be a spin-glass transition. No major effect, however, was seen. If τ_S were to change by 10%, values of J_c would change by factors of 2. No changes on this order are visible on the $J^{1/2}$-versus-T curve (open triangles) of Fig. 4. Any changes in τ_S which occur at the spin-glass temperature must be small. This aspect of the data needs to be investigated further.

The rather close fit of the data to the theory of Makeev *et al.* at low temperatures and the theory of Kogan[18] and de Gennes[2] at high temperature is a clear sign that there is a simple statistical probability for Cooper pairs to traverse the normal barrier without breaking. No other new types of correlation among spins are needed to describe the results. The data indeed show that τ_S is very nearly independent of temperature, even though one of the samples goes through the spin-glass temperature. From the results it is clear that if a junction with a strong temperature dependence to the critical current is desired one should make a thick, clean barrier. If a junction with a nearly-temperature-independent critical current is desired, one should heavily dope a thin junction with magnetic impurities.

IV. CONCLUSIONS

Critical-current densities have been measured for Pb-$Ag_{1-x-y}Mn_xAl_y$-Pb junctions over the temperature range $0.4<T<7.2$ K for a wide range of magnetic scattering rates. The I-V characteristics for these junctions show sharp reversible features without any voltage steps or hysteresis. Observed diffraction patterns indicate the junctions are of high quality and are not self-field-limited for a critical-current density of less than 80 A/cm^2 where W/λ_J is smaller than about 4. With minor modification the theory of Makeev *et al.* describes the J_c-versus-T data for all temperatures below $T/T_c \simeq 0.6$. This, in turn, implies that the order parameter at the S-N interface is independent of T in this temperature range, as required by the theory. For T near T_c, J_c was found to be proportional to $(1-T/T_{cS})^2$ as predicted by de Gennes theory.[2] The τ_s's derived from the slope of the $J_c^{1/2}$-versus-T plot scale well with the Mn concentration and are consistent with those obtained from fits to the low-temperature data. The data for 1.5-at. % Mn sample provide a spin-flip–scattering rate of about 2.95×10^{-13} sec^{-l}. Expressed in terms of the depairing temperature, this is about 8.3 K.

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