Superconducting and Magnetic Properties of Nb/Pd$_{1-x}$Fe$_x$/Nb Triple Layers

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The superconducting and magnetic properties of Nb/Pd$_{1-x}$Fe$_x$/Nb triple layers with constant Nb layer thickness $d_{Nb} = 200$ Å and different interlayer thicknesses $3$ Å $\leq d_{PdFe} < 80$ Å are investigated. The thickness dependence of the magnetization and of the superconducting transition temperature shows that for small iron concentration $x$ the Pd$_{1-x}$Fe$_x$ layer is likely to be in the paramagnetic state for very thin films whereas ferromagnetic order is established for $x \geq 0.13$. The parallel critical field $B_{c2}(T)$ exhibits a crossover from two-dimensional (2D) behavior where the Nb films are coupled across the interlayer, towards a 2D behavior of decoupled Nb films with increasing $d_{PdFe}$ and/or $x$. This 2D-2D crossover allows a determination of the penetration depth $\xi_F$ of Cooper pairs into the Pd$_{1-x}$Fe$_x$ layer as a function of $x$. For samples with a ferromagnetic interlayer $\xi_F$ is found to be independent of $x$.

PACS numbers: 74.60.Ge, 74.80.Dm, 75.70.Ak

1. INTRODUCTION

The proximity effect of a superconductor ($S$) in contact with a ferromagnet ($F$) has attracted considerable new interest, since an oscillatory behavior of the Cooper pair amplitude in the ferromagnet was predicted theoretically in $S/F$ multilayers. Due to the exchange field in the ferromagnet, the pair-breaking parameter is complex and causes a spatial modulation of the superconducting order parameter in the ferromagnetic interlayer. For certain thicknesses $d_F$ of the ferromagnetic layer the phase of the order parameter changes across the barrier (so-called $\pi$-junction) which gives rise to an enhanced transition temperature $T_c$. This, for instance, should show up in a nonmonotonic dependence of $T_c(d_F)$. Further theoretical work has shown that these anomalies should also occur in $S/F$ bilayers. Several experimental studies have been performed to search for the appearance of $\pi$-coupling in $S/F$ multilayers and triple layers. However, up to now none of the investigations have revealed unambiguously a nonmonotonic behavior due to a $\pi$-coupling mechanism. Although the measurements on sputtered Nb/Gd multilayers and triple layers have been interpreted in terms of this mechanism, the loss of ferromagnetic order at thin interlayer thicknesses or a magnetically "dead" interface region can also result in a nonmonotonic behavior of $T_c(d_F)$. Furthermore, the magnitude of the electron mean-free path $l$ in $S$ and $F$, the interface transparency, and spin-orbit scattering must be taken into account. A prominent parameter which enters all of the present theories is the characteristic complex decay constant $k_F$ describing the decay of the pair amplitude $F_F$ in the $F$ layer along the surface normal $x$, i.e. $F_F \propto \exp(-k_F x)$. The real part of $k_F$ defines the exponential decay of the envelope of $F_F$, i.e. the penetration depth of Cooper pairs in the $F$ layer, whereas the imaginary part defines oscillations of $F_F$. In the theory of Radović et al. these two length scales turn out to be identical, $(\text{Im} k_F)^{-1} = (\text{Re} k_F)^{-1} = \xi_F/2$, with the characteristic length $\xi_F$ defined as $\xi_F = \sqrt{\hbar D_F/\Sigma}$. $D_F$ is the electronic diffusion constant in $F$ and $2\Sigma$ is the splitting of the spin-up and spin-down conduction bands by the exchange interaction. However, the penetration depth and the oscillation period of $F_F$ can be different for small electron mean free paths and strong spin-orbit scattering. In recent experiments which have been mainly discussed in frame of the theory by Radović et al. the length $\xi_F$ often serves as an adjustable parameter. In this paper, we will focus on the upper critical field of triple layers and show that $\xi_F$ can be determined from a crossover in the parallel upper critical field of Nb/Pd$_{1-x}$Fe$_x$/Nb triple layers rather than use it as a free parameter whose value depends on the theoretical model employed. This crossover from two-dimensional (2D) behavior of the whole triple layer to 2D behavior of each Nb film individually, can only be observed for a thickness $d_F$ smaller than a critical thickness $d_c$, which will be identified as $\xi_F$, see below. The influence of different ferromagnetic materials on the superconducting properties of $S/F$ multilayers has been studied previously by Koorevvar et al. where the emphasis was put on the critical thickness of the superconducting layers which are decoupled by thick ferromagnetic interlayers. In contrast, the present work focuses on the influence of the ferromagnetic layer thickness in $S/F/S$ triple layers with superconducting layers of constant thickness. We will furthermore show that the analysis of the superconducting properties allows access to the magnetic properties in thin ferromagnetic films. The ferromagnetic behavior of Pd$_{1-x}$Fe$_x$ alloys has been studied in great detail, in particular the Pd-rich alloys. In Pd$_{1-x}$Fe$_x$, the Curie temperature $T_C$ can be changed over a wide range of concentration $x$. For small $x$, the polarization of the Pd conduction band around each localized Fe moment gives rise to a "giant moment" of 13 - 16 $\mu_B$ per Fe impurity. In addition, the Fe-Pd exchange interaction leads to an indirect ferromagnetic Fe-Fe interaction. In bulk alloys, ferromagnetism persists down to a concentration of about $x \approx 10^{-4}$ (Ref. [17]) and $T_C$ increases monotonically with increasing $x$. Furthermore, with increasing $x$ the electronic structure gradually changes from a localized to a
more itinerant behavior and direct Fe-Fe interactions become important. In the present work we use the large variability in $T_C$ and interaction strength with $x$ to tune the $S/F$ coupling.

II. EXPERIMENTAL

Nb single layers and Nb/Pd$_{1-x}$Fe$_x$/Nb triple layers were grown in an ultra-high vacuum system by e-beam evaporation onto in-situ cleaned Al$_2$O$_3$ (1120) substrates at room temperature. A set of eight samples with different $d_{PdFe}$ was prepared during a single evaporation process. For the triple layers, a 200-Å Nb film was deposited first onto all substrates of one set. The thicknesses were measured with a quartz-crystal monitor. During the subsequent simultaneous evaporation of Pd and Fe from two different crucibles the computer-controlled sample shutter was opened stepwise to expose the samples one after another to the Pd and Fe beams. Finally, a second 200-Å Nb layer was deposited on all samples. The relative error of the Fe concentration estimated from the variation of the evaporation rates during the process was less than 10%. Symmetrical $\theta/2\theta$ scans were taken on some samples with a thick Pd$_{1-x}$Fe$_x$ interlayer ($d_{PdFe} > 30$ Å) using a standard X-ray powder diffractometer with Cu-K$_\alpha$ radiation. The scans indicate oriented growth of bcc-Nb (110) and fcc-PdFe (111) for $x \leq 0.4$ or bcc-Fe for $x = 1$ along the surface normal. The measured lattice parameters agree within 10% with data of 1000-Å rf-sputtered Pd$_{1-x}$Fe$_x$ films.

The electrical resistivity was measured in a He$^4$ cryostat with a conventional four-point probe using spring-loaded needles in magnetic fields up to 5 T. The superconducting critical temperature $T_c$ was determined from the midpoint of the resistive transition. The transition width $\Delta T_c$, estimated from the difference in temperature at 10% and 90% of the transition, was typically $\Delta T_c \leq 40$ mK for all samples. The upper critical magnetic field $B_{c2}(T)$ was determined from the midpoint of the transition measured by ramping up the magnetic field until 90% of the transition was accomplished and ramping down again. The temperature during this sweep was held constant within typically 2 mK. The critical-field transition width is defined as the field difference at 10% and 90% of the transition. The magnetic properties were investigated by SQUID magnetometry for temperatures 2-300 K and in magnetic fields up to 5 Tesla with the field oriented parallel to the sample surface.

III. RESULTS

A. Magnetic Properties

Fig. 1 shows the dc-susceptibility $\chi(T)$ for one triple layer measured in the zero-field-cooled and field-cooled modes. The sharp decrease of the signal at low $T$ is due to the superconducting transition. The Curie temperature $T_C$ was estimated from the extrapolation to $M(T \to T_C) = 0$. $T_C$ is listed in Table I for some samples together with the values for bulk Pd$_{1-x}$Fe$_x$ alloys ($d_{PdFe} = \infty$). The ferromagnetic order was further checked on several samples of different concentration and Pd$_{1-x}$Fe$_x$ thickness by performing hysteresis loops $M(H)$ at $T = 8$ K (Fig. 1, inset).

The magnetic properties of the films change with thickness and concentration. Table I clearly shows that $T_C$ decreases with decreasing $x$ when samples of almost equal $d_{PdFe}$ are compared, as expected from the $T_C(x)$-dependence in the respective bulk alloys. Furthermore, $T_C$ decreases with decreasing $d_{PdFe}$ for fixed concentration, e.g. $x = 0.20$. This is possibly due to finite-size effects where the thickness dependence of the Curie temperature is described by $T_C \propto d_{PdFe}^{-\lambda}$ and the exponent $\lambda$ depends on the dimensionality and universality class of the system. For $x = 0.05$ the magnetic signal was very weak (not shown). From the susceptibility data a $T_C \approx 60$ K was estimated. For $x = 0.01$ a magnetic signal could not be detected. However, the magnetic behavior of these samples can be inferred from the investigation of the superconducting properties as will be discussed below. The magnetic moment per atom $\mu_{exp}$ was determined from the saturation magnetization (Table I), albeit with a large error for small $d_{PdFe}$. The average moment increases with concentration $x$ for samples with roughly equal thickness, in accordance with the concentration dependence $T_C(x)$ in bulk alloys. However, the measured values are smaller than those of bulk samples ($\mu_{exp}/\mu_{bulk} < 1$, Table I). This could be due to the existence of a magnetically "dead" layer at the Pd$_{1-x}$Fe$_x$/Nb interface as reported previously for Fe/Nb/Fe triple layers. This case, the measured val-
TABLE I. Magnetic properties of Nb/Pd$_{1-x}$Fe$_x$/Nb triple layers

| $x$ | $d_{PdFe}$ ($\AA$) | $T_c$ (K) | $\mu_{exp}(\mu_0)$ | $\mu_{exp}/\mu_{bulk}$ | Ref. |
|-----|------------------|----------|------------------|-------------------|-----|
| 0.05 | 18               | 60       | 0.17 ± 0.1       | 0.57 ± 0.3        | this work |
|      | $\infty$         |          | 0.3              |                   | 2   |
| 0.13 | 15               | 150      | 0.2 ± 0.1        | 0.3 ± 0.2         | 1   |
|      | $\infty$         |          | 0.06             |                   | 2   |
| 0.20 | 9                | 70       | 0.13 ± 0.1       | 0.14 ± 0.1        | this work |
| 12   | 175              |          | 0.1 ± 0.1        | 0.1 ± 0.1         | this work |
| 17   | $>300$           | 0.96 ± 0.24 | 1.05 ± 0.4      |                   | this work |
|      | $\infty$         | 0.91     | 1                |                   | 3   |

ues can be described by $\mu_{exp}/\mu_{bulk} = 1 - d_0/d$, where $d_0$ is the thickness of the nonmagnetic layer. From the data of Table I a thickness $d_0 \approx 8 \AA$ is estimated, i.e. 4 Å on either side of the Pd$_{1-x}$Fe$_x$ layer. The latter is smaller than the interface thickness of $\approx 7$ Å between Nb and pure Fe.

B. Upper Critical Magnetic Field of Nb Single Films

Single Nb films with various thicknesses were investigated in order to check that the parallel critical field of Nb/Pd$_{1-x}$Fe$_x$/Nb triple layers is not determined by the occurrence of surface superconductivity. Before presenting the results, we briefly summarize the usual behavior of $B_{c2}(T)$ in dependence of the orientation of the magnetic field and of the dimensionality of the sample. In general, the perpendicular critical magnetic field of superconducting films of thickness $d$ obeys a linear temperature dependence, i.e. three-dimensional (3D) behavior, below $T_c$,

$$B_{c2\perp}(T) = \frac{\phi_0}{2\pi\xi_0} (1 - T/T_c)$$

(1)

because the sample dimensions are much larger than the temperature dependent Ginzburg-Landau coherence length parallel to the film plane, $\xi_0(T) = \xi_0/\sqrt{1 - T/T_c}$, with $\xi_0 = \xi(0)$.

In the parallel orientation, $B_{c2\parallel}(T)$ can be described by a similar expression where $\xi_0 = \xi_0(0)$ is replaced by $\xi_0(\perp)$. Even in isotropic superconductors such as Nb, a difference in $\xi_0$ and $\xi_\perp$ may occur because of an anisotropic microstructure. If just below $T_c$, the perpendicular coherence length $\xi_\perp$ is larger than the thickness for very thin films, $\xi_\perp(T) >> d$, the temperature dependence of $B_{c2\parallel}$ is described by the Tinkham expression for two-dimensional (2D) superconductors,

$$B_{c2\parallel}(T) = \frac{\sqrt{12}\phi_0}{2\pi\xi_0 d} \sqrt{1 - T/T_c}.$$

(2)

Apart from a factor $\sqrt{12}/\pi$ the same result is obtained by using a Ginzburg-Landau approach for dirty and anisotropic superconductors, as has been done by Schneider and Locquet to describe the overall temperature dependence of $B_{c2\parallel}(T)$. At lower temperatures $\xi_\perp(T)$ can become smaller than the film thickness and the 3D behavior is recovered. Hence, because of the temperature dependence of $\xi(T)$ a dimensional crossover from 2D to 3D behavior should occur in $B_{c2\parallel}(T)$ with decreasing $T$. However, for sufficiently thick films this regime will not be entered due to the onset of surface superconductivity which occurs for $1.84\xi_\perp(T) < d$, i.e. when the diameter of a vortex $\approx 2\xi(T)$ is smaller than the film thickness.

In a field decreasing from well above $B_{c2\parallel}$, nucleation of superconducting regions will start near the surface leading to a superconducting sheath for fields $B_{c2\parallel} < B < B_{c3\parallel} = 1.69 B_{c2\parallel}$. Generalizing this to anisotropic superconductors one obtains a linear behavior

$$B_{c3\parallel}(T) = \frac{1.69\phi_0}{2\pi\xi_0} \xi_\perp (1 - T/T_c).$$

(3)

Therefore, in superconducting films a dimensional crossover from 2D (Eq. 2) to surface superconductivity (Eq. 3) can occur as a function of temperature and thickness in the parallel critical field.

Fig. 2a shows the temperature dependence of $B_{c2\perp}$ for single films with different thickness $d_{Nb}$. All films show a linear dependence for $T > 0.75$ characteristic for 3D behavior (Eq. 1). For lower temperatures, the data points of films with $d_{Nb} \geq 275$ Å deviate from the linear dependence with a concomitant increase in the transition width. $\Delta T_c$ is indicated by vertical bars. This behavior is presumably caused by thermally activated flux creep, which is more likely to occur in samples with a lower concentration of pinning centers. This is the case for thicker films which have a lower resistivity (Table II), i.e. a lower concentration of defects.

The parallel critical field $B_{c2\parallel}(T)$ is shown in Fig. 2b. For $T > 0.7$ all samples show a square-root dependence of $B_{c2\parallel}$, i.e. 2D behavior (Eq. 2), indicating that $\xi_\perp (T)$ is larger than the thickness $d_{Nb}$. For films with $d_{Nb} \leq 275$ Å the 2D behavior survives down to the lowest temperature. In contrast, in thicker films with $d_{Nb} \geq 400$ Å, where below a certain temperature $T^*$ the nucleation center moves from the center of the film to the surface, the temperature dependence of $B_{c2\parallel}$ changes from square-root to linear behavior due to the onset of surface superconductivity. The reduced perpendicular and parallel critical fields, i.e. $\epsilon = B_{c2\perp}(T)d^2\pi/2\phi_0$ and $h = B_{c2\parallel}(T)d^2\pi/2\phi_0$, are plotted in Fig. 3 with $T$ as an implicit parameter. For small $h$ the data follow a simple line $\epsilon = 0.33h^2$ in agreement with the theoretical prediction for the 2D regime according to Saint-James and de Gennes.

In films with $d \geq 400$ Å a change to a linear behavior due to the superconducting surface sheath is observed for $h > 2.2$, e.g. $\epsilon \approx 1.1h - 0.82$ for the 460-Å film. A behavior $\epsilon = (\xi_{0\perp}/1.695\xi_0)h$ is expected when the anisotropy of the coherence length
(Eqs. 1 and 3) is taken into account. The observed offset from a strict proportionality $\epsilon \propto h$ reflects the slightly superlinear $t$ dependence of $B_{c2\perp}$. We neglect this fact for the following qualitative discussion. The crossover between the two regimes at $h \approx 2.2$ corresponds to the reduced temperature $t^* = T^*/T_c$ marked by arrows in Fig. 2b. We obtain an anisotropy ratio $\xi_{0\perp}/\xi_{0\parallel}$ about 2 from the linear regime. With $\xi_{0\parallel}$ determined from $B_{c2\parallel}$, the coherence length $\xi_{0\parallel}$ can be derived (see Table II).

The fact that $\xi_{0\perp}$ is larger than $\xi_{0\parallel}$ can be explained by the anisotropic microstructure of the film, with columns perpendicular to the surface in addition to fine equiaxed grains, giving rise to an anisotropic electron diffusivity. Alternatively, in the dirty limit an average coherence length $\xi_0$ can be determined from electronic mean free path $\ell_0$, which was calculated from the residual resistivity $\rho$ using $\rho l = 3.75 \times 10^{-16} \Omega \text{m}^2$ and $\xi_0 = 1.33$ Å. It is reassuring that these values lie between the values of $\xi_{0\parallel}$ and $\xi_{0\perp}$ (see Table II). We conclude that for temperatures $T \geq 1.5$ K investigated here, Nb films with $d_{\text{Nb}} \leq 275$ Å show only 2D behavior in the parallel critical field without the occurrence of surface superconductivity, whereas in thicker films $B_{c2\parallel}(T)$ changes due to the onset of surface superconductivity. In this case, $\xi_{0\perp}$ can be measured. This is important for the following discussion of Nb/Pd$_{1-x}$Fe$_x$/Nb triple layers with $d_{\text{Nb}} = 200$ Å. Moreover, we note that in Nb/Pd$_{1-x}$Fe$_x$/Nb triple layers surface superconductivity is likely to occur only at even larger thicknesses due to the pairbreaking at the S/F interface.

### Table II. Superconducting parameters of Nb films

| $d_{\text{Nb}}$ (Å) | $\rho (\mu \Omega \text{cm})$ | $l$ (Å) | $\xi_{0\perp}$ (Å) | $\xi_{0\parallel}$ (Å) | $\xi_{0\perp}$ (Å) | $\xi_{0\perp}/\xi_{0\parallel}$ |
|---------------------|------------------------|--------|-------------------|-------------------|------------------|-----------------------------|
| 100                 | 15.35                  | 24     | 79                | 90                |                  |                             |
| 200                 | 11.86                  | 32     | 90                | 91                |                  |                             |
| 275                 | 8.0                    | 47     | 107               | 107               |                  |                             |
| 400                 | 5.47                   | 69     | 126               | 126               | 104              | 182                        |
| 460                 | 5.50                   | 68     | 126               | 126               | 103              | 210                        |

### C. Upper Critical Magnetic Field of Nb/Pd$_{1-x}$Fe$_x$/Nb Triple Layers

The perpendicular critical magnetic field for some selected samples of different $d_{\text{PdFe}}$ and $x$ is shown in Fig. 3. In all cases, an almost linear $T$ dependence suggesting 3D behavior is observed. Deviations at lower temperatures can be attributed to an increasing transition width in magnetic field which aggravates the precise determination of $B_{c2\perp}$. 

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**FIG. 2.** (a) Perpendicular critical magnetic field $B_{c2\perp}$ vs. reduced temperature $t = T/T_c$ for Nb single films of different thickness. Solid lines indicate the linear temperature dependence near $t = 1$. The error bars do not exceed the symbol size for $t \geq 0.7$. (b) Parallel critical field $B_{c2\parallel}(t)$. Arrows indicate the temperature $t^*$ below which surface superconductivity comes into play. Solid and dashed lines show the square-root and linear behavior above and below $t^*$, respectively.

**FIG. 3.** Plot of the scaled perpendicular critical field $\epsilon = B_{c2\perp}(\pi d^2)/(2\phi_0)$ vs. the scaled parallel critical field $\eta = B_{c2\parallel}(\pi d^2)/(2\phi_0)$. Solid and dashed lines indicate the experimental behavior for the 460-Å film $\epsilon \propto 0.33 h^2$ and $\eta \propto 1.1 h$, respectively. The transition takes place around $h \approx 2.2$ marked by arrow. Dashed-dotted line indicates $\epsilon = \eta/1.695$ without anisotropy ($\xi_{0\parallel} = \xi_{0\perp}$). Horizontal and vertical bars indicate the transition widths.
FIG. 4. Perpendicular critical magnetic field $B_{c2\perp}$ vs. temperature $T$ for Nb/Pd$_{1-x}$Fe$_x$/Nb triple layers of different thickness $d_{PdFe}$ and concentration $x$. Solid lines indicate the linear temperature dependence near $T_c$.

FIG. 5. Parallel critical magnetic field $B_{c2\parallel}$ vs. temperature $T$ for Pd$_{1-x}$Fe$_x$ triple layers of different thickness $d_{PdFe}$ and concentration $x$. For $x = 0.4$ and $d_{PdFe} = 6$ Å the solid lines indicate the different square-root behavior at temperatures below and above the 2D-2D crossover temperature $T_0$ marked by arrow.

The temperature dependence of the parallel critical field $B_{c2\parallel}$ is shown in Fig. 5. Two kinds of behavior are observed:

1. For thin interlayer thickness and/or low iron concentration (including $x = 0$) $B_{c2\parallel}(T)$ exhibits a square-root like behavior close to $T_c$ and a second square-root like behavior of different slope at lower temperatures. This is illustrated by way of example in Fig. 4, where $B_{c2\parallel}^x(T)$ plotted vs. $T$ exhibits two linear regimes with a gradual transition around a temperature $T_0$ indicated by arrows. The change in the $T$ dependence at $T_0$ and magnetic field $B_0$ is attributed to a 2D-2D crossover, similar to the case of Pb/Ge multilayers with a limited number of bilayers. Roughly speaking, just below $T_c$ both Nb layers are coupled through the interlayer, the order parameter extends over the total sample thickness and for $\xi(T) > d_{tot}$ a 2D behavior is observed (Eq. 2). At lower temperatures the coupling is suppressed by the pair-breaking of the interlayer and the individual Nb layers give rise to a second 2D behavior in $B_{c2\parallel}$. The effect of an external magnetic field on the proximity effect was studied earlier in $S/N$ junctions. In the dirty limit the temperature dependence of the coherence length in the non-magnetic metal $N$, $\xi_N = \sqrt{D/2\pi k_B T} \sim 1/\sqrt{T}$, would suggest that the coupling of the $S$ layers increases with decreasing temperature. An increasing applied magnetic field has no effect on the overlap of both pair-amplitudes from $S$ leaking into $N$ with an exponential decay until superconductivity in $N$ suddenly breaks down at the so-called break-down field $B_0$. At $B_0$ only a small superconducting sheath at the $S/N$ interface survives, which further decreases in thickness with increasing $B$. The detailed behavior depends on the thickness $d_N$ and on the transition temperature $T_cN$ of the $N$ layer. We simply adopt these results to the investigated $S/F$ system, assuming that the exchange interaction in $F$ increases the pairbreaking but does not change the qual-
ative behavior. For a strong ferromagnetic interlayer $T_{cN}$ would be zero. Therefore, the data of samples with a thin and/or weakly ferromagnetic Pd$_{1-x}$Fe$_x$-layer can be explained in the following way: In low fields the Nb films are coupled and give rise to single-film behavior with $d_{tot} \approx 2d_{Nb} + d_{PdFe}$. When the break-down field $B_0$ is reached, superconductivity in the Pd$_{1-x}$Fe$_x$ layer is almost completely suppressed and the Nb layers decouple, showing a 2D behavior in $B_{c2\parallel}$ (More precisely, the transition in the $(T, B)$ phase diagram (Fig. 1) occurs at a point where the $B_0(T)$ line crosses the $B_{c2\parallel}(T)$ curve.) This is further confirmed by the calculated coherence lengths $\xi_{||}$ (Eq. 2) which agree well with those obtained from the $B_{c2\perp}(T)$ data if we assume $d = d_{tot}$ for coupled films ($T > T_0$) and $d = d_{Nb} = 200\,\text{Å}$ for decoupled films ($T < T_0$). The effective thickness of $S$ might be somewhat smaller due to the proximity of the ferromagnetic material. This would lead to higher values of $\xi_{||}$ and better agreement with the values determined from the $B_{c2\perp}(T)$ behavior as shown above.

(2) In samples with a large $d_{PdFe}$ and/or large $x$ the individual Nb layers are decoupled for all $T$ by the strong pairbreaking of the magnetic interlayer. In this case a single square-root behavior of $B_{c2\parallel}(T)$ is observed down to the lowest temperature (cf. Fig. 3, $x = 0.40, d_{PdFe} = 26\,\text{Å}$), similar to the case of a single 200-Å Nb film (Fig. 2b). Hence, the 2D-2D crossover depends on the interlayer thickness and the iron concentration. Fig. 3 immediately shows that for constant $x$ and increasing $d_{PdFe}$ the characteristic temperature $T_0$ shifts towards $T_c$ which in turn decreases, until the individual Nb films are decoupled for all temperatures (cf. Fig. 3, $x = 0.40$). The $T_0(x, d_{PdFe})$ dependence is seen more clearly in Fig. 4 where the reduced values $(1 - T_0/T_c)$ are plotted vs. $d_{PdFe}$ for different $x$.

The data can be separated into two regimes. For small $x < 0.05$ a gradual decrease to large $d_{PdFe}$ is seen. Moreover, for constant $d_{PdFe}$, these samples show a systematic decrease of $(1 - T_0/T_c)$ with increasing $x$. In contrast, for $x \geq 0.13$ the data seem to follow a single line with a steep decrease to $(1 - T_0/T_c) = 0$ at small $d_{PdFe}$. (Note that for $d_{PdFe} \to 0, T_0$ should be zero for all $x$.) From the intersection of the data with the abscissa a critical thickness $d_c$ can be determined in dependence of $x$. In samples with $x \leq 0.05$ this thickness was estimated by a linear extrapolation of the data. For each concentration, $d_c(x)$ is the smallest $d_{PdFe}$ for which a crossover is still observed whereas samples with $d_{PdFe} > d_c$ show a $B_{c2\parallel}(T)$ behavior of decoupled Nb films for all temperatures $T < T_c$. Thus, in samples with $d_{PdFe} = d_c$ the two pair amplitudes decaying from both layers into the $F$ layer do not overlap, i.e. $d_c/2 \approx (Re_{F})^{-1} = \xi_F/2$, Ref. 2. Therefore, in the following $d_c$ will be identified as $\xi_F$ as previously done by Koorevar et al. for V/Fe multilayers.

The concentration dependence $d_c(x)$ is plotted in the inset. Again, the two different regimes can be identified. First, $d_c$ decreases slightly with increasing $x$ but becomes almost independent of $x$ after a precipitous decrease between $x = 0.05$ and 0.13. Obviously, these two regimes are related to different pairbreaking mechanisms at the $S/F$ interface, which will be discussed later. In the following, $d_c$ will be identified as $\xi_F$.

**D. Superconducting Transition Temperature of Nb/Pd$_{1-x}$Fe$_x$/Nb Triple Layers**

The superconducting transition temperatures $T_c$ vs. interlayer thickness $d_{PdFe}$ for sample sets of different $x$ with constant $d_{Nb} = 200\,\text{Å}$ are shown in Fig. 5. The transition width corresponds to the symbol size. For pure Pd ($x = 0$) a monotonic decrease of $T_c(d_{PdFe})$ from $T_c(0) = 9\,\text{K}$ is observed. Similar behavior has been reported recently for Nb/Pd multilayers with $d_{Nb}$ which has been discussed in frame of the de Gennes-Werthamer theory. With increasing $x$, the $T_c(d_{PdFe})$ curves are systematically lowered until for $x = 0.13$ and 0.20 a non-monotonic behavior with a shallow minimum between $d_{PdFe} = 10 - 20\,\text{Å}$ is seen. For these sample sets, $T_c$ becomes independent of the interlayer thickness for large $d_{PdFe}$. Note that for $x = 0.20$ all samples very likely have a ferromagnetic interlayer since already for $d_{PdFe} = 9\,\text{Å}$ a ferromagnetic hysteresis loop and a Curie temperature $T_C \approx 70\,\text{K}$ were measured (Table 1). Therefore, a nonmonotonic behavior caused by the establishment of long-range ferromagnetic order beyond a certain interlayer thickness, as observed in Fe/Nb/Fe triple layers, can be ruled out.

The thickness where the minimum occurs for $x = 0.13$ and 0.20 is equal to the value of $d_c$ determined from the critical-field behavior. This indicates that once the
films are decoupled the transition temperature becomes independent of the thickness of the ferromagnetic interlayer. This has also been found in V/Fe multilayers for different $x$. Solid lines serve as guide to the eye. Dashed lines show extrapolations to $T_c(d_{\text{PdFe}} = 0)$. The inset shows a fit by the theory of Radović et al. (dashed-dotted line) to the data for $x = 0.20$, see text for further details.

FIG. 8. Superconducting transition temperature $T_c$ vs. interlayer thickness $d_{\text{PdFe}}$ for different $x$. Solid lines serve as guide to the eye. Dashed lines show extrapolations to $T_c(d_{\text{PdFe}} = 0)$. The inset shows a fit by the theory of Radović et al. (dashed-dotted line) to the data for $x = 0.20$, see text for further details.

IV. DISCUSSION

Obviously, the concentration dependence of the critical-field behavior and of the transition temperature can be separated into two different scenarios. For low concentrations $x \leq 0.05$ the reduced crossover temperature ($1 - T_0/T_c$) as well as $T_c$ gradually decrease with increasing $d_{\text{PdFe}}$. For these samples the critical thickness $d_c$ is large ($d_c \approx 100$ Å) and the individual Nb layers are coupled via the Pd$_{1-x}$Fe$_x$ interlayer. From the magnetic measurements it is not possible to decide unambiguously whether these small-$d_{\text{PdFe}}$ samples exhibit long-range ferromagnetic order. In contrast, for $x \geq 0.13$ the magnetic measurements show definitely that the interlayer is ferromagnetically ordered. In this case a clear transition from coupled to decoupled behavior in $B_{c2}(T)$, where $T_c$ becomes independent of $d_{\text{PdFe}}$, is found. This transition appears at a thickness $d_c \approx 12$ Å, much lower than for $x \leq 0.05$. This crossover allows an experimental determination of the characteristic penetration depth of Cooper pairs in the ferromagnetic layer, $\xi_F$, a quantity that is usually not accessible in experiment. In the theory of Radović et al.  the minimum of the $T_c(d_{\text{PdFe}})$ curve appears around $d_c \approx \xi_F$. Therefore, the critical-field measurements may serve as an independent method for the determination of $\xi_F$. First, we consider the two extreme cases of a pure Fe interlayer ($x = 1$) and a pure Pd interlayer ($x = 0$). For a pure Fe interlayer we can calculate the parameter $I = 4hD_F/\xi_F^2$ from the measured $\xi_F \approx 12$ Å. For the estimation of $D_F = v_F l/3 = 2.7$ cm$^2$/s we take $l \approx \xi_F$ as the electronic mean free path and a Fermi velocity $v_F = 6.9 \times 10^7$ cm/s for sp electronic fluids. This yields $I \approx 0.5$ eV as a lower limit in fair agreement with $I \approx 1$ eV for bulk iron. For a pure Pd interlayer, which is presumably not in a ferromagnetic state, at least not for $T > 1.5$ K, the application of the theory is questionable. An upper limit of $I \approx 80$ meV can be estimated as before, which is unrealistically low, even for the $4d - 4d$ exchange interaction in pure Pd. However, spin fluctuations or "paramagnons" with a characteristic energy of $E = 21$ meV are possibly pairbreaking in Pd. Moreover, tunneling experiments on Pd-Pb sandwiches have shown that Pd is not gapless down to a thickness of 500 Å which indicates that the pairbreaking is much weaker than by
magnetic impurities and in magnetic fields thus leading to a large $\xi_F$. Hence, for the extreme cases of $x = 0$ and $x = 1$ the experimentally determined values of $\xi_F$ can be reasonably explained by pairbreaking due to the presence of spin fluctuations and by the exchange interaction, respectively. Concerning the samples with an alloy interlayer the interesting point is that $d_c$, i.e. $\xi_F$, is more or less independent of concentration for $0.13 \leq x \leq 1$. Band-structure calculations of ordered fcc Fe-Pd alloys show that the average exchange splitting decreases by no more than a factor of two when the Fe content is successively reduced from Fe$_3$Pd to FePd. The decrease is basically due to the weaker 4$d - 4d$ exchange interaction compared to the 3$d - 3d$ interaction. Furthermore, the Fermi velocity of Pd is smaller than of Fe, $v_F^{\text{Pd}} = 2 \times 10^7$ cm/s. This suggests that with decreasing $x$ the accompanying decrease of $I$ is more or less compensated by a decrease of $D_F$. Besides, $\xi_F$ depends only weakly on $D_F/I$, viz. $\propto \sqrt{D_F/I}$. This explains why $d_c \approx$ constant for $0.13 \leq x \leq 1$. For low concentrations $x \leq 0.05$ the critical thickness is much larger. Since in this regime the interlayer is likely to be in the paramagnetic state the reduction of $T_c$ is presumably due to the well-known pair-breaking caused by spin-flip scattering first investigated in the classical work by Hauser et al. This is further corroborated by the gradual increase of $\xi_F$ with decreasing $x$ (Fig. 7). Hence, the large change of $\xi_F$ around $x \approx 0.1$ marks the transition from a paramagnetic to a ferromagnetic interlayer, similar to the previous reports on Nb/Gd and Nb/Fe systems, where such a behavior was found in dependence of the thickness of the $F$ layer. This large difference between $\xi_F$ in both regimes demonstrates that the pairbreaking by the exchange interaction is much stronger than by spin-flip scattering. Hence, the concentration dependence of $d_c$ extracted from the superconducting measurements can be considered as a magnetic response, although the magnetic properties of very diluted samples could not be measured directly.

V. CONCLUSION

The occurrence of a 2D-2D crossover in the parallel critical field of Nb/Pd$_{1-x}$Fe$_x$/Nb triple layers allows a determination of the characteristic penetration depth of Cooper pairs $\xi_F$ in the $F$ layer. With increasing $x$, this length changes abruptly in a small concentration interval due to a transition from a paramagnetic to a ferromagnetic interlayer leading to a change in the underlying pairbreaking mechanism while it is found to be more or less independent of $x$ in the ferromagnetic regime. It is reassuring that the strong difference in $\xi_F$ between $x = 0.05$ and 0.13 is reflected in the different $T_c(d_{\text{pair}})$ behavior. Although the present work presents a step forward in identifying the requirements for possible $\pi$-junctions in $S/F$ layers or multilayers, a decisive experimental test for this possibility must await phsyse sensitive measurements such as the Josephson effect.

ACKNOWLEDGMENTS

We thank M. Kelemen for performing the SQUID measurements. This work was partly supported by the Deutsche Forschungsgemeinschaft through Graduiertenkolleg "Kollektive Phänomene im Festkörper".

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