PHYSICAL REVIEW B **80**, 214523 (2009)

Spin screening effect in superconductor/ferromagnet thin film heterostructures studied using nuclear magnetic resonance

R. I. Salikhov,* N. N. Garif'yanov, and I. A. Garifullin
Zavoisky Physical-Technical Institute, 420029 Kazan, Russia

L. R. Tagirov
Kazan State University, 42008 Kazan, Russia

K. Westerholt and H. Zabel
Institut für Experimentalphysik/Festkörperphysik, Ruhr-Universität Bochum, D-44780 Bochum, Germany

(Received 10 July 2009; revised manuscript received 27 October 2009; published 21 December 2009)

Using NMR spectroscopy of the ^{51}V nuclei in the superconducting state of Ni/V/Ni and $Pd_{1-x}Fe_x/V/Pd_{1-x}Fe_x$ trilayers we reported in a recent letter an experimental observation of the spin screening effect [R. I. Salikhov, I. A. Garifullin, N. N. Garif'yanov, L. R. Tagirov, K. Theis-Bröhl, K. Westerholt, and H. Zabel, Phys. Rev. Lett. **102**, 087003 (2009)]. This effect, which designates the formation of a spin polarization in the superconducting state, was predicted previously by Bergeret *et al.* [F. S. Bergeret, A. F. Volkov, and K. B. Efetov, EPL **66**, 111 (2004); Phys. Rev. B **69**, 174504 (2004)]. Here, we extend our earlier experiments by varying the thickness of the superconducting V layer and by applying the magnetic field not only perpendicular to the film plane as in the previous experiments, but also in the parallel direction. For the latter geometry, which for experimental reasons is difficult to realize, the film is in the vortex-free state. This allows a direct quantitative comparison of the experimental screening effect as derived from a characteristic distortion of the high-field wing of the resonance line in the superconducting state and the theoretical model calculations. We derive a reasonable agreement between theory and experiment, confirming the spin screening effect in the superconductor.

DOI: 10.1103/PhysRevB.80.214523 PACS number(s): 74.45.+c, 74.25.Nf, 74.78.Fk

I. INTRODUCTION

The mutual influence of magnetism and superconductivity in superconductor/ferromagnet (S/F) nanofabricated thin film heterostructures has been an exciting topic in solid-state physics during the last 15 years (see, e.g., reviews 1–5). As emphasized frequently in these reviews, there are interesting theoretical predictions still waiting for unambiguous experimental verification. These are, e.g., the generation of the long-range triplet superconductivity in the F layer or the so-called spin screening effect (also sometimes called the inverse proximity effect), which designates a spin polarization in the superconducting layer close to the S/F interface. In the theory of the spin screening effect[6,7] it is shown that in the S layer a spin polarization with a direction opposite to the ferromagnetic magnetization of the F layer develops below the superconducting transition temperature T_c. This implies, in simple terms, that the superconducting layer becomes ferromagnetically polarized with a magnetization direction antiparallel to the ferromagnetic layer below T_c.

Qualitatively the physical origin of this spin screening effect can easily be understood. Let us consider a S/F bilayer with the F layer being thin compared to the superconducting coherence length (or the Cooper pair size) in the F layer, ξ_F. Due to the exchange field, the conduction-electron spins in the F layer are polarized in one direction predominantly. These electrons have their Cooper partners deep in the S layer and thus, due to the superconducting correlations, a spin polarization is induced in the S layer. This spin polarization has a sign opposite to the spin polarization of the conduction electrons in the F layer and a characteristic penetration depth into the S layer of the order of the coherence length in the superconducting layer ξ_s.

For a real S/F bilayer the amplitude of the magnetization induced by the spin screening effect in the S layer is expected to be very small; thus, for an experimental proof of the spin screening effect one needs a method which can sensitively probe small changes in the spin polarization in the S layer below T_c. Principally one can investigate the penetration profile of the spin polarization using the technique of low-energy muon spin rotation (see, e.g., Ref. 8). However, estimates show that the detection of the effect is on the verge of sensitivity of this technique. The induced spin polarization in the superconducting state lead to a change in the spin susceptibility of the conduction electrons upon the superconducting transition. This spin susceptibility, on the other hand, is one of the physical reasons for the Knight shift of the nuclear magnetic resonance (NMR) line in metals. Thus, in NMR the spin screening effect should manifest itself as a decrease in the Knight shift on the transition to the superconducting state. Actually, as we have discussed in our previous paper,[9] using high sensitivity NMR we were able to observe the distortion of the high-field wing of the ^{51}V NMR signal in $Pd_{1-x}Fe_x/V/Pd_{1-x}Fe_x$ and Ni/V/Ni trilayers and comparing it on a qualitative level with a simple model calculation we came to the conclusion that this distortion is caused by the spin screening effect. Another possibility to detect the spin screening effect was demonstrated recently by Xia *et al.*[10] who used the optical polar Kerr effect on Al/Co-Pd bilayers and observed a small change in the Kerr rotation below T_c of Al.

These two recent papers stimulated new theoretical and experimental efforts. Linder *et al.*[11] pointed out that in the case of a dominating triplet component of Cooper pairs penetrating the F layer, the spin polarization in the S layer could reverse its direction, i.e., it should be parallel to the magnetization of the F layer. Long-range triplet pairing in the F layer is expected theoretically if the magnetization at the interfaces is inhomogeneous, e.g., in the case of an in-plane domain wall at the S/F interface.[11] Asulin *et al.*[12] investigated the effect of the induced magnetization on the density of states (DOS) of the high-T_c superconductor $YBa_2Cu_3O_7$ (YBaCuO) with *d*-wave superconductivity covered by $SrRuO_3$ (SRO) ferromagnetic islands. Using tunneling spectroscopy they measured the DOS in YBaCuO films in the vicinity of the ferromagnetic SRO islands. Surprisingly, they found that the distance of penetration of the magnetic ordering into the S layers is one order of magnitude larger than the Cooper pair size. The authors interpreted their results assuming the importance of another relevant length scale in the proximity problem, namely, the spin diffusion length.

For our present investigation of the spin screening effect by NMR the choice of an appropriate F/S material combination is of primary importance. It is desirable that the S-layer material has a strong NMR signal with a small linewidth, a suitable superconducting transition temperature T_c and a high-quality interface with the F material. In addition, there should be an appreciable change in the Knight shift at the transition to the superconducting state. Among the elemental superconductors, Pb, Nb, and V appear to be possible candidates.[13,14] However, only V fulfills the condition of a high interface quality with epitaxial growth of Fe on V and high interface transparency for the electrons.[15,16] At the same time the early results of Noer and Knight[17] indicated that the Knight shift for V does not change markedly at T_c, which would render V unsuitable for the present study. However, as we have shown recently, in pure V the Knight shift definitely changes below T_c,[18] very similar to pure Nb,[19] which has a similar electronic structure.

In order to obtain a measurable spin polarization caused by the spin screening effect, the S-layer thickness in the S/F bilayer should be comparable to ξ_s, because the perturbation of the spin susceptibility in the S layer is expected at a distance of the order ξ_s from the S/F interface only. On the other hand, at the thickness of the S layer smaller than $2\xi_s$, superconductivity usually vanishes (see, e.g., Ref. 20). Therefore, the S-layer thickness in the S/F bilayer is limited to about $2\xi_s$. Usually[16] for our V films $\xi_s \simeq 10$ nm, implying that the number of V nuclei involved in the resonance will be extremely small. In order to increase the number of V nuclei subjected to the spin screening effect we used trilayer samples F/S/F (i.e., one S layer between two F layers) for our present investigation. Nevertheless, conventional NMR spectrometers encounter serious sensitivity problems with this small sample volume and we had to develop a supersensitive NMR technique operating in a continuous mode to reach the necessary sensitivity.

In the experimental section below we carefully study the behavior of the NMR signal of Ni/V/Ni trilayers for different thicknesses in perpendicular as well as parallel orientation of the dc magnetic field relative to the film plane. In our earlier investigation of Ni/V/Ni and $Pd_{1-x}Fe_x/V/Pd_{1-x}Fe_x$ we only studied the NMR signal for the perpendicular orientation. We compare the NMR signal in the normal and superconducting states and also study in detail a single V film as a reference.

TABLE I. Experimental parameters of all samples for the present study: S1 is the single V layer, S2 is the $Pd_{0.98}Fe_{0.02}/V/Pd_{0.98}Fe_{0.02}$ trilayer, S3 is the $Pd_{0.97}Fe_{0.03}/V/Pd_{0.97}Fe_{0.03}$ trilayer, and S4 and S5 are the Ni/V/Ni trilayers with thicknesses of the V layer of 44 and 70 nm, respectively. Given are the thickness of the V layer d_V, the roughness parameter σ, the superconducting transition temperature T_c, the residual resistivity ratio RRR, the electron mean free path in the V layer l, and the superconducting coherence length ξ_s. The thickness of the magnetic layers is 3 nm for all trilayer samples.

Sample	d_V (nm)	σ (nm)	T_c (K)	RRR	l (nm)	ξ_s (nm)
S1	30	0.3	4.65	11	15	14
S2	36	1.3	3.02	4.6	5	8
S3	42	1.3	3.55	6	7	10
S4	44	1.6	4.05	4.4	5	8
S5	70	0.8	4.4	8.2	10.5	11.6

The paper is organized as follows. Section II provides a brief outline of the sample preparation and characterization. This section also contains a description of our NMR setup. Results of the NMR measurements and their analysis are given in Sec. III. In Sec. IV the experimental results are discussed in the framework of the theoretical model by Bergeret *et al.*[6] Finally, the main results are summarized in Sec. V.

II. EXPERIMENTAL DETAILS

A. Sample preparation and characterization

We have prepared a number of F/S/F trilayers with V as the superconducting layer and either Ni or an alloy $Pd_{1-x}Fe_x$ as the ferromagnetic layers (see Table I). All layers were grown on single-crystalline MgO(001) substrates with dimensions of 10×10 mm^2 by molecular beam epitaxy (MBE). In a first step the MgO substrate was preheated at 600 °C at a pressure of 10^{-6} mbar for 1 h in the load lock chamber of the MBE system. Then it was transferred to the growth chamber, where the substrate was heated to 1000 °C at a base pressure of 5×10^{-10} mbar for 5 min. All layers were grown in the growth chamber with a base pressure below 5×10^{-10} mbar and at a substrate temperature of 300 °C. This growth temperature provides an optimized compromise between crystal quality and interdiffusion at the interfaces. For V, Ni, and Pd we used electron beam evaporation and growth rates of 0.15, 0.03, and 0.05 nm/s, respectively. The $Pd_{1-x}Fe_x$ alloy layers were produced by coevaporation of Pd and Fe. Fe was evaporated from an effusion cell with an evaporation rate depending on the desired concentration of Fe in the alloy. To prevent oxidation, all samples were capped by 2-nm-thick Pd layers. *In situ* reflection high-energy electron diffraction during the growth process revealed smooth layer growth of all layers.

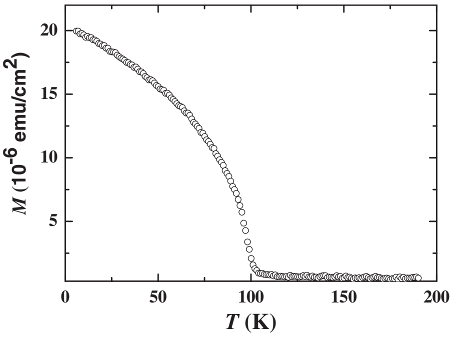

FIG. 1. Magnetization versus temperature for a $Pd_{0.97}Fe_{0.03}/V/Pd_{0.97}Fe_{0.03}$ trilayer (sample S3) measured in a magnetic field of 100 G.

The thickness and the quality of the films were characterized by conventional small-angle x-ray reflectivity. Well-resolved Kiessig fringes from the total layer thickness were clearly observed. Fits using the modified Parratt formalism[21,22] yield the thickness of the V layers, d_V, and the interface roughness parameter σ given in Table I. Due to the fact that the structural quality of V grown directly on MgO is much higher than for the growth on the Ni and $Pd_{1-x}Fe_x$ surfaces, the σ values for the single V layer and the trilayer systems are quite different (see Table I, column 3).

The concentrations of Fe in the $Pd_{1-x}Fe_x$ alloy layers were refined with the data for $T_{Curie}=f(x)$ taken from the literature.[23] From the temperature dependence of the magnetization measured in a magnetic field of 100 G using a superconducting quantum interference device magnetometer (Fig. 1), we derived ferromagnetic Curie temperatures of $T_{Curie}\simeq 75$ and 100 K for two samples S2 and S3 in Table I. For these Curie temperatures we estimate Fe concentrations x of 0.02 and 0.03, respectively.

The upper critical magnetic fields $B_{c2}^{\parallel}$ and $B_{c2}^{\perp}$ for the magnetic field direction parallel and perpendicular to the film plane were measured resistively by standard four-point dc technique (Fig. 2). The superconducting transition temperature T_c for the samples in Table I is between 3 and 4.7 K (see the fourth column of Table I). From the residual resistivity ratio $\text{RRR}=R(300\text{ K})/R(5\text{ K})$ (fifth column of Table I) we can determine the specific residual resistivity ρ_0 using the phonon contribution to the specific resistivity for vanadium, $\rho_{phon}(300\text{ K})=18.2\ \mu\Omega$ cm. Following Lazar *et al.*[20] with the Pippard relations,[24] we get $\rho_0 l=2.5\times10^{-6}\ \mu\Omega\text{ cm}^2$ and we can calculate the mean free path l of the conduction electrons (sixth column of Table I). The BCS coherence length for V is $\xi_0=44$ nm. A comparison of l and ξ_0 implies that the superconducting parameters of our samples are closer to the "dirty" limit ($l\ll\xi_0$) than to the "clean" limit ($l\gg\xi_0$). In the dirty limit $\xi_s=\sqrt{\xi_0 l/3.4}$ holds, which is given in the last column of Table I.

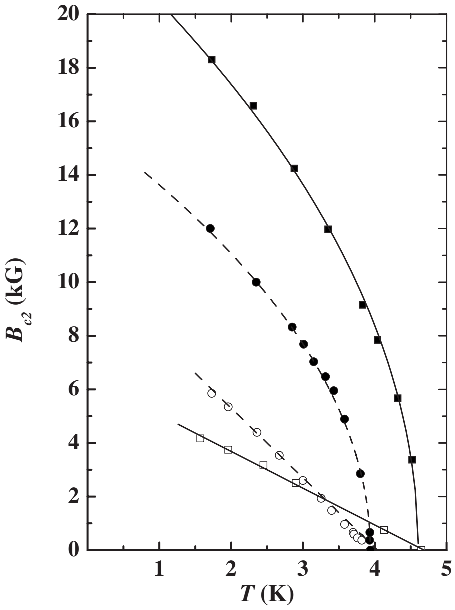

FIG. 2. Upper critical magnetic field for the single V layer (sample S1) (squares) and for the Ni/V/Ni trilayer (sample S4) (circles). Closed symbols correspond to the field direction parallel to the film plane; the opened symbols correspond to the perpendicular orientation.

B. NMR spectrometer

We have built a continuous-wave NMR spectrometer operating at the frequency of about 5.5 MHz (Ref. 25) and based on a self-oscillating detector (see, e.g., Ref. 26). Using the metal semiconductor field effect transistor (MESFET) CF739 capable of operating at temperatures below 4 K, we were able to immerse the high-frequency generator into the liquid helium in close vicinity to the pick-up coil. This strongly reduces the thermal noise and excludes losses in the line connecting the pick-up coil with the generator.

Our spectrometer uses a conventional phase sensitive detector with a magnetic field modulation frequency of 917 Hz. In order to keep the signal-to-noise ratio high, it is important to minimize the ac signal generated by the modulating field in the pick-up coil. Therefore, we precisely adjusted the axes of the signal and modulation coils perpendicular to each other at room temperature. At liquid-helium temperature the adjustment becomes worse due to thermal expansion. To minimize this effect we mounted both coils on a single block made of beech wood with a very small thermal-expansion coefficient.

Since the gyromagnetic ratios for the Cu and V nuclei are very similar, the resonator coil as well as the magnetic field modulation coils was wound of high-purity Ag wire. At liquid-helium temperatures, the resonance circuit has a high Q value, which also considerably enhances the NMR spectrometer sensitivity. The input impedance of the MESFET is high enough to prevent the Q value of the tank circuit from being lowered. The generator output was connected to the lock-in amplifier PAR-5209. For the generation of the dc magnetic field we used the magnet system of the Bruker EPR spectrometer ER-418, which includes a field sweep option and a stabilization by a Hall unit. Precision measurements of the magnetic field were performed by a NMR gaussmeter whose NMR sensor was always in a strictly fixed position. The experimental error in the measurement of the magnetic field including its inhomogeneity in the operating range (4×10^{-5}) did not exceed 0.5 G.

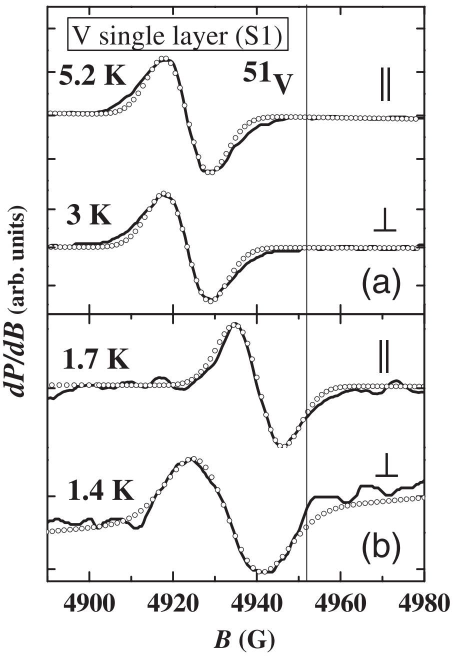

FIG. 3. NMR spectra for the single V layer (sample S1) in the (a) normal and (b) superconducting states for parallel (∥) and perpendicular (⊥) orientations of dc magnetic field. The NMR spectra are fitted by the Gaussian line shape (circles). Here, and in the following figures the vertical line shows the NMR line position for ^{51}V nuclei in an insulator.

III. RESULTS OF THE NMR MEASUREMENTS AND THEIR ANALYSIS

Measurements of the NMR signal of the ^{51}V nuclei were performed in the temperature range between 1.4 and 5.2 K. Since the operating frequencies are slightly different for different samples, in order to compare the resonance line positions directly, all data were recalculated to the same frequency ν, in our case to $\nu=5542.3$ kHz. For all samples the signal-to-noise ratio does not exceed a factor of 3. Therefore, we accumulated signals from at least 20–30 sweeps of the magnetic field taken during 2 min each. Figures 3–5 contain data for the normal and the superconducting states.

A. Normal state

In Fig. 3(a) we show the NMR signals for the single V layer (sample S1) in the normal state for the parallel and perpendicular orientations of the dc magnetic field relative to the film plane. The resonance line shape is well described by the derivative of a Gaussian absorption curve. Fitting this theoretical curve to the experimental spectra we can determine the resonance line position with an absolute accuracy better than 0.5 G. For the resonance linewidth (the peak-to-peak distance of the absorption line derivative) we get a value of $\Delta B=11.2$ G. The resonance field of $B_0^n=4923.1$ G is shifted by $\delta B=29.1$ G relative to its position in an insulator (4952.2 G for ^{51}V). Thus, for the Knight shift in the normal state, which is defined as the ratio of the NMR line shift relative to its position in an insulator, we get $(0.59\pm0.01)\%$, in good agreement with the value measured previously.[17,18] The NMR line shape in the superconducting state is discussed in the next paragraph.

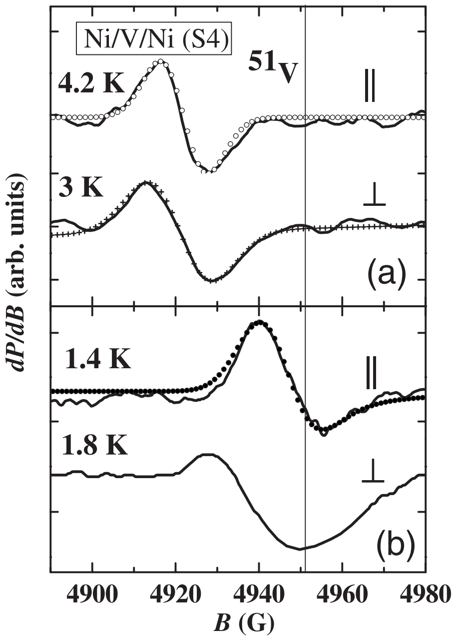

FIG. 4. NMR spectra for Ni/V/Ni trilayer (sample S4) in the (a) normal and (b) superconducting states for the parallel (∥) and perpendicular (⊥) orientations of the field. The NMR spectrum for the normal state in the parallel orientation is fitted by the Gaussian line shape (open circles), and in the perpendicular orientation by the Gaussian line shape taking the demagnetizing field from the F layers into account (crosses). The fit for the superconducting state in parallel orientation takes the spin screening effect with $B_m=15$ G [see Eqs. (1) and (2)] into account (closed circles).

Figure 4(a) displays the NMR signals for a Ni/V/Ni trilayer (sample S4) in the normal state for both orientations of the magnetic field. For the field direction parallel to the film plane the resonance line position and the linewidth coincide nicely with that observed for the single V layer [Fig. 3(a)]. For the perpendicular orientation of the field the NMR signal is shifted toward lower magnetic fields by 3 G and the line shape appears slightly distorted (the low-field wing has a smaller amplitude than the high-field wing). These observations are not surprising since for the field directed parallel to the film plane the magnetization of the F layer lies in plane and the demagnetizing field acting on the V layer is negligible. For the perpendicular orientation the demagnetizing field from the F layers is nonzero. We numerically estimated this dipolar field and obtained that this field slightly shifts the resonance line to the low-field side and causes some line broadening with the degree of broadening comparable to the shift. As a result, the amplitude of the low-field wing of the resonance line becomes slightly smaller than the amplitude of the high-field wing, just as observed in the experiment. The calculated resonance line for the perpendicular direction is shown in Fig. 4(a) by circles and it is obvious that there is satisfactory agreement with the experimental resonance lines.

Figure 5 shows the NMR signals in the normal state for $Pd_{0.98}Fe_{0.02}/V/Pd_{0.98}Fe_{0.02}$ [Fig. 5(a)] and

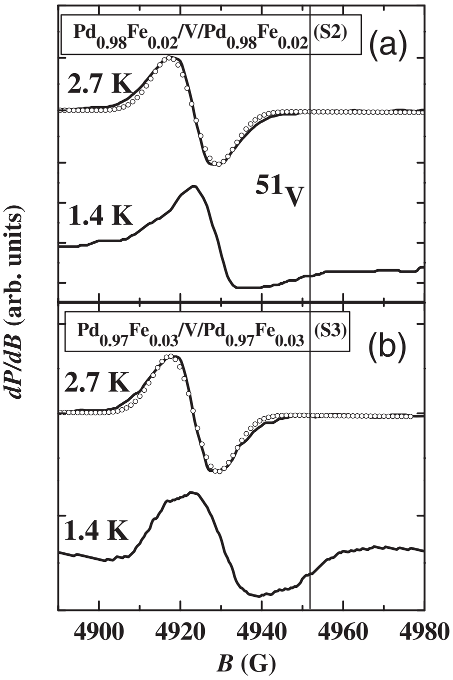

FIG. 5. NMR spectra (a) for the $Pd_{0.98}Fe_{0.02}/V/Pd_{0.98}Fe_{0.02}$ trilayer (sample S2) in the normal ($T=2.7$ K) and superconducting ($T=1.4$ K) states and (b) for the $Pd_{0.97}Fe_{0.03}/V/Pd_{0.97}Fe_{0.03}$ trilayer (sample S3) in the normal ($T=2.7$ K) and superconducting ($T=1.4$ K) states. All data refer to the perpendicular magnetic field direction. The NMR spectra for the normal state are simulated with the Gaussian line shape (circles).

$Pd_{0.97}Fe_{0.03}/V/Pd_{0.97}Fe_{0.03}$ [Fig. 5(b)] for the field perpendicular to the film plane. The resonance line shape for these samples in the normal state is well described by the derivative of a Gaussian absorption curve, in contrast to the case of Ni/V/Ni samples in the perpendicular orientation just discussed. The reason is that the magnetization of the strongly diluted $Pd_{1-x}Fe_x$ alloy is very small and therefore the demagnetizing field has virtually no influence.

B. Superconducting state

During the first stage of our NMR experiments in the superconducting state we encountered serious sensitivity problems. Aside from a nonlinear drift of the zero line when sweeping the dc magnetic field, we observed regular noise with an amplitude exceeding the noise of the spectrometer. The nonlinear drift of the zero line when measuring a superconducting sample is a well-known phenomenon, which we have observed already in our NMR study of bulk V samples.[18] It is caused by the nonlinear dependence of the surface impedance of the superconductor on the magnetic field and is usually registered together with the resonance signal (see, e.g., Ref. 27). Regular noise from a superconducting sample is present for both magnetic field directions, but at the first stage of our NMR study for the field direction parallel to the film plane this noise was much larger than for the perpendicular direction.[28]

In Fig. 3(b) the NMR spectrum for the single V layer (sample S1) below T_c for both field orientations is depicted. Compared to the normal state [Fig. 3(a)] the resonance line is shifted toward higher magnetic fields and definitely broadened in case of the perpendicular orientation ($\Delta B=15.5$ G). After aligning the sample position with respect to the magnetic field with a high precision, and after zero-field cooling of the sample, the noise level and the drift of the base line were reduced considerably thus enabling the observation of high-quality NMR spectra for the parallel field orientation, too. The reason for this drastic improvement is not completely clear; however, we suppose that the main reason is that after the zero-field cooling and with the magnetic field axis exactly parallel to the sample plane, the superconducting V layer is in a vortex-free state. Burger *et al.*[29] studied systematically the dependence of the critical field of thin films of type II superconductors on the parameters d_s, λ, and ξ_s and provided experimental evidence that for the case of $d_s \simeq \lambda$ and $B \ll B_{c2}$ and the field parallel to the film plane the films are in the vortex-free state. The thickness of the V layers, d_V, in our samples is small compared to the magnetic penetration depth $\lambda \sim 50$ nm and, as seen in Fig. 2, $B_{c2}^{par} \simeq 3B_0$ holds, thus justifying our assumption that the samples are in the vortex-free state.

Vanadium is a type II superconductor and for the perpendicular orientation the V film is in the vortex state. The broadening and shift of the NMR line upon the transition to the superconducting state is caused by the inhomogeneous magnetic field distribution in the vortex state. The NMR line shape in the mixed state of type II superconductors is determined by the convolution of the normal-state line shape and the singular distribution of the magnetic field in the vortex state (see, e.g., Refs. 19, 30, and 31). Brandt[32] argued that in order to observe this particular distribution of the local field in the vortex state one needs a pin-free ellipsoidal sample made of high-purity single-crystalline material with the Ginzburg-Landau parameter $\kappa \sim 1$ (for example, ultrapure Nb). In real V samples pinning of the vortices is unavoidable. For our samples with $\kappa \simeq 3-4$ the pinning forces lead to a transformation of the singular field distribution to a Gaussian shape[32] with a width estimated as $\delta B_v \sim (B_{c2}-B_0)/2\kappa^2$. With $B_{c2} \simeq 5000$ G and $B_0=4920$ G this gives $\delta B_v \sim 3.5$ G. If the NMR line shape in the normal state is Gaussian, then in the superconducting state it should keep its Gaussian shape with some additional broadening δB_v as estimated above. This is just what we have observed in our experimental spectrum for the single vanadium film (sample S1). [See the evolution of the NMR linewidth from Fig. 3(a) (normal state, $\Delta B=11.2$ G) to Fig. 3(b) (superconducting state with $\Delta B=15.5$ G)]. Upon the transition to the superconducting state the line shape does not change markedly, the resonance field increases up to $B_0^s=4943$ G, and an additional Gaussian broadening $\delta B_v^{exp} \simeq 4.3$ G is observed. The vortex motions and their depinning lead to the appearance of the regular noise in Fig. 3(b).

For the single V film [Fig. 3(b)] in the parallel orientation, we first note that, similar to the perpendicular orientation, the NMR line shifts to higher fields compared to the normal state. However, in contrast to the perpendicular orientation, the NMR linewidth does not markedly differ from the normal state. This supports our assumption above that the broadening in the perpendicular orientation is caused by the pres-

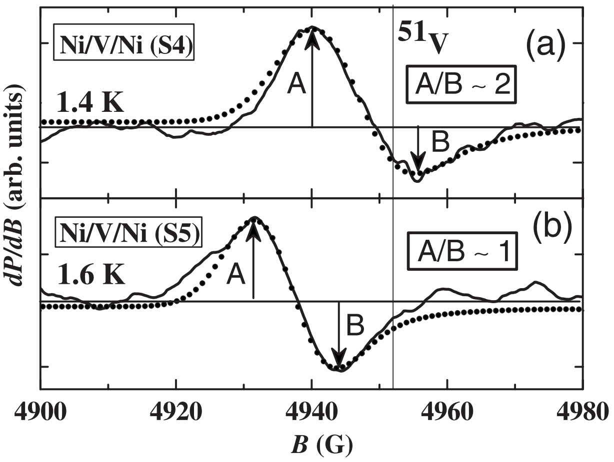

FIG. 6. NMR spectra for Ni/V/Ni trilayers (samples S4 with $d_V=44$ nm and S5 with $d_V=70$ nm) in the superconducting state (parallel magnetic field). The theoretical fits take the spin screening effect with $B_m=15$ G [see Eqs. (1) and (2)] into account (closed circles).

ence of vortices. For the parallel orientation vortices are absent; thus, the magnetic field inside the V layer decays exponentially from both surfaces with the decay length given by the magnetic penetration depth λ. Numerical calculations show that in our case for $d_V \sim 30$ nm and $\lambda \sim 50$ nm the inhomogeneity of the magnetic field distribution virtually does not influence the NMR line width, because the magnetic field is strongly inhomogeneous only in the close vicinity of the film surface. The convolution of the field distribution with a Gaussian line shape in this case leads to the shift of the resonance line by less than 1 G and to a small distortion of the resonance line wings only.

Figure 4(b) shows the NMR spectra for Ni/V/Ni trilayer (sample S4) in the superconducting state for both orientations of magnetic field. Similar to the case of the single V layer we observe a shift of the resonance line to higher magnetic fields. At the same time, however, the line shape for both field directions is markedly changed with the high-field wing of the NMR line strongly distorted. As we have discussed above and shown, for the example, for a single V layer, the transition to the superconducting state should not change the Gaussian shape of the NMR line. For our F/S/F trilayers we have a Ginzburg-Landau parameters even larger than for the single V layer ($\kappa \simeq 4-5$) and we expect stronger pinning forces because of the sandwiching of the S layer between two F layers.[33] The same anomalous change in the NMR line shape we also observe for the NMR spectra in $\mathrm{Pd}_{1-x}\mathrm{Fe}_x/\mathrm{V}/\mathrm{Pd}_{1-x}\mathrm{Fe}_x$ trilayers with $x=0.02$ (sample S2) and 0.03 (sample S3) in the superconducting state (Fig. 5).

We also studied the evolution of the NMR line shape with increasing S-layer thickness for Ni/V/Ni trilayer samples (Fig. 6). One sees that the distortion of the high-field wing of the resonance line has an obvious trend to disappear with increasing V layer thickness.

IV. DISCUSSION OF THE SPIN SCREENING EFFECT

The central result of our present study is that the NMR line shape of the F/S/F trilayers definitely changes on the transition to the superconducting state. This does hold for the field orientation perpendicular to the sample plane as well as for the parallel orientation. Compared to the Gaussian line shape for the single vanadium layer (for both the normal and the superconducting states) and F/V/F trilayers in the normal state the line shape for F/S/F trilayers in the superconducting state is asymmetric with reduced amplitude of the high-field peak B (see, e.g., Fig. 6). The line shape for sample S4 in the superconducting state is reminiscent of the classical calculation by Bloembergen[34] for the NMR line shape in the metallic samples with a thickness d comparable to the electrodynamic skin depth δ. The line-shape asymmetry parameter A/B (the ratio of low-field peak height A to the high-field peak height B) varies from 1.0 for fully transparent films ($d \ll \delta$) to 2.55 for a half space ($d \gg \delta$). This distortion results from electrodynamic admixture of the dispersion component of the dynamic magnetic susceptibility to the absorption component. For a metallic half space, the detected NMR signal is a one-to-one mixture of the absorption and dispersion, and the asymmetry parameter reaches its maximum magnitude $A/B=2.55$. At our NMR frequency of 5.5 MHz, the skin depth is about 50 μm. Thus, in the normal state samples with a total thickness on the order of 40–70 nm are completely transparent for the radio frequency radiation; hence, no electrodynamic distortion of the NMR line shape is expected. The absence of electrodynamic distortion of the line shape ($A/B \sim 1$) upon the superconducting transition for the single V film and for the samples with the thick V layer [sample S5 in Fig. 6(b)] provides additional experimental evidence that the asymmetric NMR line shape observed in F/V/F trilayers in the superconducting state is not a consequence of electrodynamic screening.

A further possible origin of a distorted NMR line could be an inhomogeneous distribution of the quadrupole splitting at the MgO/V or the Ni/V interfaces due to the lattice mismatch. However, this type of distortion should already appear in the normal state of the V layer, which is clearly not the case (see Figs. 3–5). Thus, this mechanism can also be ruled out.

A third conventional explanation for an asymmetric line shape in the superconducting state could be the local field distribution in the vortex state. We have argued above that the pinning forces lead to a transformation of the singular field distribution of the local fields in the vortex state to a Gaussian one. This means that if the NMR line shape in the normal state is Gaussian then in the superconducting state it should keep its Gaussian shape with some additional broadening. This is just what we observe for the single V layer (Fig. 3). This fact again shows that the presence of vortices, too, cannot explain the characteristic distortion of the line shape which we observe in the trilayers.

One important experimental feature of the distortion is that it disappears with increasing V thickness, clearly indicating that there is a mechanism determining the line shape below T_c only acting in the vicinity of the interfaces at a distance on the order of 10–20 nm. When the superconducting vanadium layer is thick, the NMR signal from the unperturbed core of the film dominates in the NMR response, and the symmetry of the line shape is being restored: the asymmetry parameter A/B approaches 1. Recollecting all findings concerning the NMR line distortion we are led to the conclusion that the spin screening effect as discussed in the Introduction is the most plausible mechanism giving rise to the NMR line distortion observed experimentally.

According to the model of the spin screening effect,[6] spin-polarized electrons from the interfacial region penetrate into the superconducting layer. By means of the hyperfine interaction this spin polarization induces a local field B_{loc} on the V nuclei with a direction opposite to the external magnetic field (we suppose that the conduction-electron spin polarization in the ferromagnetic layer is in the direction of the applied field) and the NMR resonance field shifts to higher fields accordingly.

In order to calculate the NMR line shape quantitatively, one must take the spatial distribution of the spin polarization in the superconducting layer into account. The induced spin polarization in the superconductor which is proportional to the local magnetic field B_{loc} decays exponentially with the distance x from both F/S interfaces,

$$P(x) \sim B_{loc} = B_m \cosh(k_s x), \tag{1}$$

where the x axis is perpendicular to the S/F interface and $x=0$ corresponds to the center of the superconducting layer, $k_s = 1/\xi_s$, and B_m is the value of the local field at the S/F interfaces. The local field distribution,

$$F(B) = \frac{1}{d}\int_0^d dx\ \delta[B - B_{loc}(x)], \tag{2}$$

has to be convoluted with the unperturbed NMR Gaussian line shape derived from the normal-state NMR line above T_c.

Fitting the NMR line shape with the local field modified by the spin screening effect is straightforward for the case of the parallel field direction since in this case the film is in the vortex-free state and there are no complications due to the inhomogeneous local field distribution in the vortex state. As seen in Fig. 3, the NMR line for the single V layer in the parallel orientation of the sample simply shifts to higher fields without any broadening below T_c.

The fits taking the spin screening effect into account [Figs. 4(b) and 6] show a reasonable agreement with the experimental line shape. We obtain a parameter $B_m \simeq 15$ G, which represents the maximum shift of the resonance line for nuclei in close vicinity of the S/F interface. The resonance field value was taken as a free parameter in the fit.

We next want to try a quantitative comparison of B_m resulting from the fit and the corresponding theoretical model of the spin screening effect by Bergeret *et al.*[6] Within this model the local magnetic field producing the polarization of conduction electrons at the interface is given by

$$B(\pm d_s/2) = \alpha 4\pi M_F (d_f/\xi_s). \tag{3}$$

Here, α denotes the part of the magnetization of the ferromagnet caused by the conduction electrons. Using the saturation magnetization of Ni, $M_F=515$ G, and supposing that metallic Ni is an ideal itinerant ferromagnet ($\alpha \simeq 1$), we get $B(d_s/2) \simeq 3$ kG. This field produces the polarization of the conduction electrons in the superconducting layer and via the contact interaction shifts the NMR line. One should remember that the origin of this shift is the same as the origin of the Knight shift where the polarization of the conduction electrons produced by an external magnetic field causes the line shift.

Figure 3 shows that in the parallel orientation of the single V film the NMR resonance field in the normal state is $B_0^n = 4923.1$ G. The shift of the resonance line relative to the position in an insulator (4952.2 G) is $\delta B = 29.1$ G. In the superconducting state the resonance field is $B_0^s = 4943$ G (Fig. 3). As mentioned above, our estimation shows that the diamagnetism of the film due to the Meissner effect contributes less than 1 G to the shift of the resonance field. Therefore, the shift of the NMR line by $B_0^s - B_0^n \simeq 20$ G at the transition into the superconducting state is solely due to the change of the Knight shift, i.e., the change in the electron polarization at the V core produced by an external magnetic field of about 5 kG. This provides a suitable reference for the calculation of the parameter $B_m = 15$ G, the spin screening parameter which we have fitted above in Figs. 4(b) and 6. In the theory of the spin screening effect B_m is caused by the induction of Ni at the interfaces $B(\pm d_s/2) \simeq 3$ kG. With the relation between the induction and change in the Knight shift in the superconducting state (5 kG gives a shift $\delta B \simeq 20$ G), the theory predicts $B_m \simeq 12$ G, in good agreement with $B_m = 15$ G derived experimentally. One should bear in mind that the theoretical model is rather crude and does not take into account the complicated electronic band structure of V and Ni and parameters such as the roughness and the finite transparency of the Ni/V interface.

For the perpendicular orientation of the magnetic field, one must take into account the local field distribution due to the spin screening effect as well as the inhomogeneous field distribution due to the vortex state. For the numerical calculations one would need the map of the flux distribution at the film surface, which is hard to get, especially if there is vortex pinning. Therefore, we did not try to fit these spectra quantitatively and just present qualitatively the tendency of the broadening of the high-field wing of the NMR line for the S2, S3, and S4 trilayer samples.

V. SUMMARY AND CONCLUSIONS

We have prepared F/S/F trilayer samples with ferromagnetic $Pd_{1-x}Fe_x$ or Ni layers and a superconducting V layer and performed a systematic NMR study of the ^{51}V nuclei for temperatures above and below the superconducting transition temperature T_c. In an extension of our previous investigation we show the NMR for the magnetic field direction oriented parallel to the film plane. In a control experiment we also studied in detail the resonance line shape and the Knight shift of a single V film of comparable thickness without contact to ferromagnetic layers.

We find first qualitative and then quantitative manifestations of the spin screening effect in the superconducting state, as evidenced by a characteristic asymmetry of the NMR line shape below T_c. The asymmetry is similar for both magnetic field directions, which is important since with the field parallel to the sample plane there are no vortices in the V film and thus it can be ruled out that a complicated vortex distribution giving rise to field inhomogeneities can cause the distortion of the NMR line. We obtained a good agreement between experimental and theoretical resonance line shapes calculated taking the spin screening effect into account. We have also shown that the spin screening effect disappears with increasing the V-layer thickness, which is also in accordance with the theory of the spin screening effect that affects the NMR line only for nuclei close to the interfaces. The character of the NMR line distortion below T_c, its dependence on the field direction, and the thickness of the superconducting layer give further strong evidence for the mechanism called spin screening (or inverse proximity) effect in the theoretical papers.[6,7]

ACKNOWLEDGMENTS

We are grateful to Anatoly F. Volkov for his help in the estimation of the local magnetic field arising due to the spin screening effect and general discussion of the obtained results. This work was supported by the Deutsche Forschungsgemeinschaft within the SFB 491 and by the Russian Foundation for Basic Research [Projects No. 08-02-00098 (experiment) and No. 07-02-00963 (theory)].

---

*ruslan_salikhov@yahoo.com

[1] I. A. Garifullin, J. Magn. Magn. Mater. **240**, 571 (2002).

[2] Yu. A. Izyumov, Yu. N. Proshin, and M. Khusainov, Usp. Fiz. Nauk **172**, 113 (2002) [Phys. Usp. **45**, 109 (2002)].

[3] A. I. Buzdin, Rev. Mod. Phys. **77**, 935 (2005).

[4] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, Rev. Mod. Phys. **77**, 1321 (2005).

[5] K. B. Efetov, I. A. Garifullin, A. F. Volkov, and K. Westerholt, in *Magnetic Heterostructures: Advances and Perspectives in Spinstructures and Spintransport*, Springer Series Tracts in Modern Physics Vol. 227, edited by H. Zabel and S. D. Bader (Springer, New York, 2007), p. 252.

[6] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, EPL **66**, 111 (2004); Phys. Rev. B **69**, 174504 (2004).

[7] M. Yu. Kharitonov, A. F. Volkov, and K. B. Efetov, Phys. Rev. B **73**, 054511 (2006).

[8] A. Suter, E. Morenzoni, R. Khasanov, H. Luetkens, T. Prokscha, and N. Garifianov, Phys. Rev. Lett. **92**, 087001 (2004).

[9] R. I. Salikhov, I. A. Garifullin, N. N. Garif'yanov, L. R. Tagirov, K. Theis-Bröhl, K. Westerholt, and H. Zabel, Phys. Rev. Lett. **102**, 087003 (2009).

[10] J. Xia, V. Shelukhin, M. Karpovski, A. Kapitulnik, and A. Palevski, Phys. Rev. Lett. **102**, 087004 (2009).

[11] J. Linder, T. Yokoyama, and A. Sudbo, Phys. Rev. B **79**, 054523 (2009).

[12] I. Asulin, O. Yuli, G. Koren, and O. Millo, Phys. Rev. B **79**, 174524 (2009).

[13] J. R. Schrieffer, *Theory of Superconductivity* (W. A. Benjamin, New York, 1964).

[14] D. E. MacLaughlin, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press, Inc., New York, 1976), Vol. 31, p. 1.

[15] P. Isberg, B. Hjörvarsson, R. Wräppling, E. B. Svedberg, and L. Hulman, Vacuum **48**, 483 (1997).

[16] I. A. Garifullin, D. A. Tikhonov, N. N. Garif'yanov, L. Lazar, Yu. V. Goryunov, S. Ya. Khlebnikov, L. R. Tagirov, K. Westerholt, and H. Zabel, Phys. Rev. B **66**, 020505(R) (2002).

[17] B. J. Noer and W. D. Knight, Rev. Mod. Phys. **36**, 177 (1964).

[18] I. A. Garifullin, N. N. Garif'yanov, R. I. Salikhov, and L. R. Tagirov, Pis'ma Zh. Eksp. Teor. Fiz. **87**, 367 (2008) [JETP Lett. **87**, 316 (2008)].

[19] D. Rossier and D. E. MacLaughlin, Phys. Kondens. Mater. **11**, 66 (1970).

[20] L. Lazar, K. Westerholt, H. Zabel, L. R. Tagirov, Yu. V. Goryunov, N. N. Garif'yanov, and I. A. Garifullin, Phys. Rev. B **61**, 3711 (2000).

[21] L. C. Parratt, Phys. Rev. **95**, 359 (1954).

[22] L. Névot and P. Croce, Rev. Phys. Appl. **15**, 761 (1980).

[23] J. A. Mydosh, J. I. Budnick, M. P. Kawatra, and S. Skalski, Phys. Rev. Lett. **21**, 1346 (1968).

[24] A. B. Pippard, Rep. Prog. Phys. **23**, 176 (1960).

[25] In order to observe the NMR in the superconducting state the resonance field should be smaller than the critical field of superconductor (see Fig. 2). For the chosen frequency the NMR of ^{51}V nuclei occurs at the magnetic field on the order of 5 kOe (see Figs. 3–6).

[26] K. J. Wilson and C. P. G. Valabhan, Meas. Sci. Technol. **1**, 458 (1990).

[27] N. E. Alekseevskii, I. A. Garifullin, B. I. Kochelaev, and E. G. Kharakhsh'yan, Pis'ma Zh. Eksp. Teor. Fiz. **18**, 323 (1973) [JETP Lett. **18**, 189 (1973)].

[28] This was the reason why in our previous paper we presented only NMR data for the perpendicular orientation.

[29] J. P. Burger, G. Deutscher, E. Guyon, and A. Martinet, Phys. Rev. **137**, A853 (1965).

[30] J.-M. Delrieu, Solid State Commun. **8**, 61 (1970).

[31] L. Dobrosavljevic, C. R. Hebd. Seances Acad. Sci. **263**, 502 (1966).

[32] E. H. Brandt, J. Low Temp. Phys. **73**, 355 (1988).

[33] E. H. Brandt, Physica B (to be published).

[34] N. Bloembergen, J. Appl. Phys. **23**, 1383 (1952).