Proximity effect in Nb/Cu/CoFe trilayers

Kyungmoon Kim, Jun Hyung Kwon, Jinho Kim, and K. Char*
School of Physics and Center for Strongly Correlated Materials Research, Seoul National University, Seoul, Korea

Hyeonjin Doh† and Han-Yong Choi
Department of Physics, Bk21 Physics Research Division, and Institute for Basic Science Research, Sung Kyun Kwan University, Suwon 440-746, Korea

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We have fabricated Nb/Cu/CoFe trilayers, varying the thickness of the Cu or CoFe layer in order to control the coupling between the superconductivity and the ferromagnetism. The T_c behavior of the Nb/Cu bilayer exhibits the well-known proximity effect of an SN bilayer. In a Nb/Cu/CoFe trilayer with the thicknesses of the Nb and CoFe layers fixed, the T_c values increase on two different scales of length as the thickness of the Cu layer increases: one with a length scale of about 2 nm when the Cu layer is less than 5 nm in thickness and the other with a length scale of about 68 nm, the normal coherence length of Cu, when the Cu layer is larger than 5 nm in thickness. Furthermore, in a similar manner to the case of Nb/CoFe, the T_c behavior of a Nb/Cu/CoFe trilayer as a function of CoFe thickness shows a small dip structure. By employing Usadel formalism, we deduced all the relevant parameters that can explain our data in a consistent manner. In spite of the better interfaces in the Nb/Cu/CoFe system, no T_c oscillation as a function of the Cu thickness was observed, in contrast to the Nb/Au/CoFe system. We will discuss the difference and similarity between the Nb/Cu/CoFe and Nb/Au/CoFe systems.

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INTRODUCTION

The proximity effect that exists between a superconductor (S) and a normal metal (N) has been well known for several decades.[1–6] When a normal metal is placed in close contact with a superconductor, there is finite possibility of finding the cooper pairs in the normal metal due to the proximity effect. Therefore, as the normal metal thickness increases, the T_c of the bilayer decreases exponentially with a characteristic length, ξ_N.[4–6] On the other hand, when a ferromagnetic metal (F) is used instead of a normal metal, the cooper pairs entering the ferromagnetic region acquire additional center of mass momentum due to the exchange field. This adds an oscillating term to the Cooper pair wave function inside the ferromagnetic region, resulting in oscillatory behavior of the T_c of the bilayers as a function of the ferromagnet thickness,[7–22] which is known as the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) picture.[23,24]

In our previous paper,[25] we inserted Au as a normal metal between a superconductor and a ferromagnet in order to study the interaction between them. In this Nb/Au/CoFe system T_c oscillation as a function of Au thickness with a length scale of about 20 nm was observed, although no satisfactory explanation for it has been found yet. In the mean time the T_c oscillation as a function of Au thickness was observed again in the epitaxial Nb/Au/Co system[26] with a much smaller length scale, about 2 nm. In this paper, instead of Au we used Cu, which is known to have good interfaces with Nb and CoFe, in the hope that a better interface would prove more effective in promoting the interaction between the Nb and CoFe layers. We will present the T_c behavior of the Nb/Cu/CoFe trilayer from our experiments as a function of the thickness of the Cu or CoFe layers. In addition, we will analyze our data quantitatively using the method based[19,28] on the Usadel formalism.[27]

EXPERIMENTAL METHOD

We prepared Nb/Cu bilayer and Nb/Cu/CoFe trilayer samples by a dc magnetron sputtering system at room temperature. We used 200 nm-oxidized Si wafers of lateral size 2×7 mm^2 as our substrates. After the sputtering chamber was evacuated to 2×10^{-8} Torr, each film was deposited using 99.999% pure argon gas at 4 mTorr. First, a Nb (99.95% pure) layer was deposited simultaneously on several substrates arranged in a line in an area of a holder where the uniformity of T_c was measured to be within 20 mK. Subsequently, Cu (99.996% pure) and $Co_{60}Fe_{40}$ (99.9% pure) layers were deposited *in situ* to prevent Nb oxidation. To reduce the experimental errors among the samples, we used a wedge-shaped deposition geometry. By this method, we were able to obtain a systematic variation in thickness the Cu and CoFe layers using the natural gradient of the sputtering rate, created by placing the substrate in an asymmetric position relative to the center of the target. As the final step, all our samples were capped by a 3 nm thick Al layer to prevent oxidation during measurement. The deposition rates of each layer were 0.299 nm/sec for Nb, 0.248 nm/sec for Cu, and 0.079 nm/sec for CoFe.

The superconducting transition temperature, T_c, of the sample was measured resistively using a standard DC 4-point method with a 0.1 mA current and determined from a resistance vs temperature (R vs T) curve using the 10% criterion. Typical rms roughnesses of the Nb single layer, Nb/Cu bilayer, and Nb/Cu/CoFe trilayer measured by an atomic force microscope were about 0.17 nm, 0.19 nm, and 0.19 nm, respectively.

RESULTS AND ANALYSIS

R vs T curves of a Nb/Cu bilayer and a Nb/Cu/CoFe trilayer near T_c with variable thicknesses of the Cu layer are

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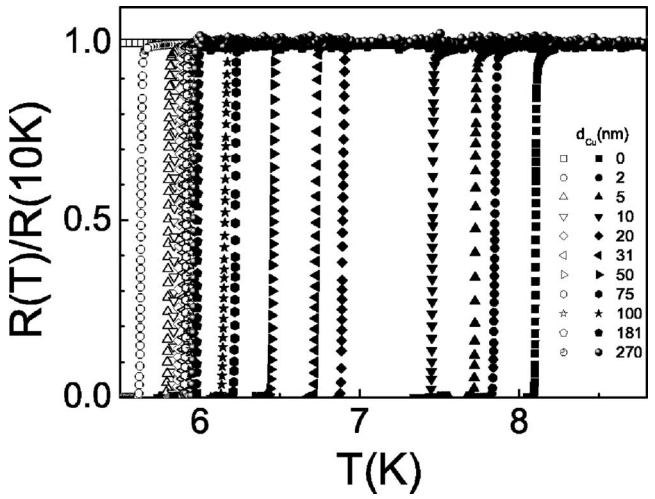

FIG. 1. Normalized R vs T curves of Nb(25.4 nm)/Cu bilayers (solid symbols) and Nb(25.4 nm)/Cu/CoFe(10 nm) trilayers (open symbols) near T_c with varying Cu thickness. The transition curves are parallel to each other for all the samples. Resistances were normalized by the value in the normal state at $T=10$ K. The T_c values were determined from the normalized R vs T curves using the 10% criterion.

presented in Fig. 1. The transition width, corresponding to the temperature difference between 10% and 90% of the normalized resistance, is about 20 mK for our samples.

As seen in Fig. 2, the T_c of the Nb/Cu bilayer with increasing d_{Cu} (thickness of Cu layer) decreases monotonically from the T_c of a Nb single layer and saturates around $d_{\mathrm{Cu}} \approx 200$ nm. In contrast, the T_c of the Nb/Cu/CoFe trilayer increases rapidly from the T_c of the Nb/CoFe bilayer as soon as the Cu layer is inserted until $d_{\mathrm{Cu}} \approx 5$ nm and then slowly approaches a limiting value around $d_{\mathrm{Cu}} \approx 200$ nm. As expected, the T_c values of the Nb/Cu bilayer and Nb/Cu/CoFe trilayer are the same above $d_{\mathrm{Cu}} \approx 200$ nm within experimental error, because the Nb layer is not affected by the CoFe layer when the Cu layer is very thick. The saturation value of the T_c of Nb/Cu/CoFe is lower than for the case of Nb/Au/CoFe, indicating that the interface resistance between Nb and Cu layers is smaller than that between Nb and Au layers. This is because the overall T_c values increase with the interface resistance of the SN boundary.

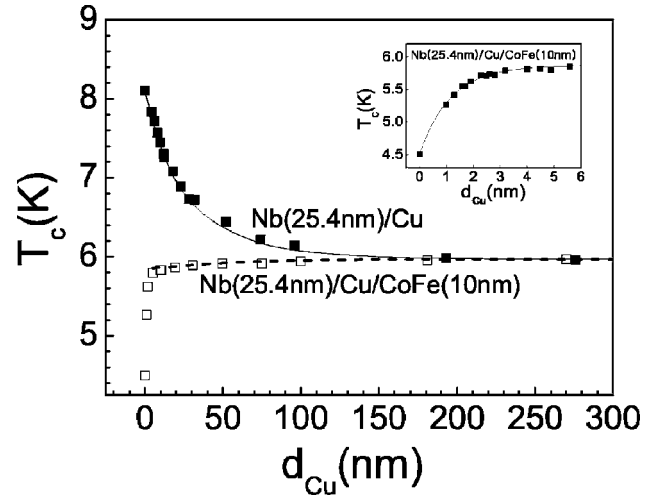

FIG. 2. T_c behavior of Nb(25.4 nm)/Cu bilayers (solid symbol) and Nb(25.4 nm)/Cu/CoFe(10 nm) trilayers (open symbol) as a function of Cu thickness. The solid (dashed) line is the result of calculation for the Nb/Cu(Nb/Cu/CoFe) system. Inset: Magnified view of T_c of Nb(25.4 nm)/Cu(d_{Cu})/CoFe(10 nm) trilayers. The solid line is a result of first order exponential decaying fit with a characteristic length of $\kappa^{-1}=2.2$ nm.

A quantitative analysis based on Usadel formalism was used to analyze the T_c behavior of SN and SNF systems in a consistent manner, and the calculation procedure was presented in our previous paper.[25] Here, the parameters necessary for the calculation are the T_{cS} (T_c of a Nb single layer), ρ (resistivity), and ξ (dirty limit coherence length) of each metal, γ_b^{SN} (the interface parameter between S and N), γ_b^{NF} (the interface parameter between N and F), and E_{ex} (the exchange energy of F). We obtained $T_{\mathrm{cS}}=8.09$ K, $\rho_{\mathrm{Nb}}=15.2\ \mu\Omega$ cm, $\rho_{\mathrm{Cu}}=1.9\ \mu\Omega$ cm, $\rho_{\mathrm{CoFe}}=14.8\ \mu\Omega$ cm, and $E_{\mathrm{ex}}=99.4$ meV from the separate experiments; T_{cS} was obtained by measuring the T_c of a Nb single layer, ρ of each metal by measuring the residual resistivity at 10 K, and E_{ex} of the CoFe layer from our previous work on Nb/CoFe bilayers.[22] In the calculation, these parameters from the separate experiments were inserted as constants and the rest of the parameters were adjusted to provide a good fit to our data.

The best fit result for the Nb/Cu bilayers was obtained with the parameters as $\xi_{\mathrm{Nb}}=7.0$ nm, $\xi_{\mathrm{Cu}}=66$ nm, and $\gamma_b^{\mathrm{SN}}=0.1$. This best fit result is represented as a solid line in Fig. 2 and is in good agreement with our data. The mean free paths of the Nb and the Cu layers can be calculated from their coherence lengths. The mean free path of the Nb is $l_{\mathrm{Nb}} \approx 1.7$ nm which is calculated by inserting the ξ_{Nb} value and $v_{\mathrm{F}}=0.56\times10^6$ m/sec (Ref. 29) into the definition of the dirty limit coherence length, $\xi_S=\sqrt{\hbar v_F l_{\mathrm{Nb}}/6\pi k_B T_{\mathrm{cS}}}$. By using the same method, we obtained $l_{\mathrm{Cu}} \approx 55.4$ nm for the mean free path of the Cu. These values satisfy the dirty limit condition and seem reasonable. The interface resistance between S and N is calculated to be $R_b^{\mathrm{SN}}A=0.13\times10^{-11}\ \Omega\ \mathrm{cm}^2$ from $\gamma_b^{\mathrm{SN}} \equiv \frac{R_b^{\mathrm{SN}}A}{\rho_N \xi_N}=0.1$ and is indeed very small, especially compared with the value of the Nb/Au case.[25]

With the same method as used in the Nb/Cu bilayer, we also calculated the T_c of a Nb/Cu(d_{Cu})/CoFe trilayer, taking the effect of two interfaces into account. We used the same parameters as used for the bilayers of Nb and Cu. No fitting parameters of ξ_{CoFe} and γ_b^{NF} could be made to fit the data of the small thickness range below $d_{\mathrm{Cu}} \approx 5$ nm where T_c increases very rapidly with respect to small variations in d_{Cu}. Except for this small thickness range, the best fit result is obtained with the parameters as $\xi_{\mathrm{CoFe}}=13.5$ nm and $\gamma_b^{\mathrm{NF}}=0.44$. A dash line in Fig. 2 shows this fitting result is in good agreement with our data. The corresponding interface resistance between N and F is $R_b^{\mathrm{NF}}A=0.88\times10^{-11}\ \Omega\ \mathrm{cm}^2$ from the equation, $\gamma_b^{\mathrm{NF}} \equiv \frac{R_b^{\mathrm{NF}}A}{\rho_F \xi_F}$.

An expanded view of the T_c behavior of Nb/Cu(d_{Cu})/CoFe trilayers in the small thickness range of Cu is shown in the inset of Fig. 2. As soon as a Cu layer is inserted, the data exhibit a rapid increase in T_c. Because this

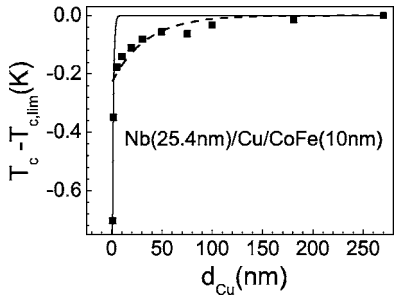

FIG. 3. Rescaled view of T_c of Nb(25.4 nm)/Cu(d_{Cu})/CoFe(10 nm) trilayers (solid square) in Fig. 2. The solid (dashed) line is a result of first order exponential decaying fit with κ^{-1} =2.2 nm(68 nm).

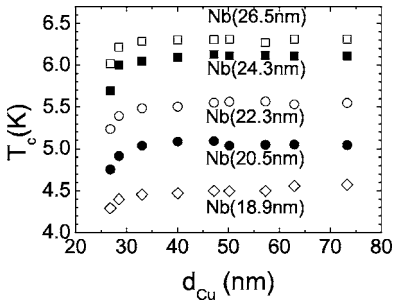

FIG. 4. T_c behavior of Nb/Cu/CoFe(10 nm) trilayers as a function of Cu thickness for various Nb thicknesses. Even for the thinnest Nb case, no clear T_c oscillation was found within our resolution.

increase cannot be understood by using the method based on Usadel formalism, we analyzed this region by employing a phenomenological equation $T_{cSNF}=T_{lim}+C^*\exp(-2d_N/\kappa^{-1*})$ similar to the de Gennes-Werthamer theory[4–6] of the SN bilayer. Unlike the bilayer case, however, the fitting parameter C^* is negative in sign. We obtained the length scale for the initial increase, $\kappa^{-1*}\approx 2.2$ nm. In the case of Au,[25] this length scale was $\kappa^{-1*}\approx 2.0$ nm.

In a similar manner, we calculated the length scale of the thickness range above $d_{Cu}\approx 5$ nm in Nb/Cu(d_{Cu})/CoFe trilayers with the phenomenological equation above. The result is $\kappa^{-1*}\approx 68$ nm, which is basically the coherence length of Cu, $\xi_{Cu}=66$ nm. Therefore, we need two length scales to explain T_c behavior in the $S/N(d_N)/F$ trilayer. One is $\kappa^{-1*}\approx 2.2$ nm for the "thin" range below $d_{Cu}\approx 5$ nm, and the other is $\kappa^{-1*}\approx 68$ nm for the "thick" range above $d_{Cu}\approx 5$ nm. Figure 3 shows these two length scales.

In Fig. 2 our data for the Nb/Cu(d_{Cu})/CoFe trilayer agree well with results calculated using Usadel formalism, in the range of $d_{Cu}\geq 5$ nm. In the calculation, we considered the interface to be a hypothetical layer of infinitesimal thickness and finite resistance. The reason why the calculation result is not in accord with our data in the range $d_{Cu}\leq 5$ nm is that, experimentally some finite distance is necessary for the full effect from the both interfaces to come into play independently. The length scale of Cu for the initial T_c increase, 2.2 nm, in the Nb/Cu(d_{Cu})/CoFe trilayer seems too large to be regarded as a necessary length for the formation of a continuous Cu layer, especially in view of the roughness of our samples, about 0.2 nm. Instead, it probably represents the length scale of the intrinsic scattering mechanism at the interfaces. This is because, in spite of the much lower vale of $\gamma_b^{SN}=0.1$ and $\gamma_b^{NF}=0.44$ in the case of Nb/Cu/CoFe, compared with $\gamma_b^{SN}=1.15$ and $\gamma_b^{NF}=0.5$ in the case of Nb/Au/CoFe,[25] the length scale for the initial T_c increase did not become smaller.

On the other hand, as a function of d_{Cu}, no T_c oscillation was observed. This is a definite contrast from the case of the Nb/Au/CoFe system, where we have seen T_c oscillation as a function of d_{Au} with a period of about 20 nm.[25] In order to confirm the absence of T_c oscillation behavior, we have reduced the Nb thickness systematically and have not observed any sign of T_c oscillation, in contrast to the Nb/Au/CoFe system, as shown in Fig. 4. It may be the larger interface parameter $\gamma_b^{SN}=1.15$ for Nb/Au, namely the large reflectance at that the Nb/Au interface, that is necessary to observe the T_c oscillation, since the corresponding interface parameter $\gamma_b^{SN}=0.1$ for Nb/Cu is much smaller.

We also studied the T_c behavior while changing the thickness of the CoFe layer in a Nb/Cu/CoFe trilayer with fixed d_{Nb} and d_{Cu}. As seen in Fig. 5, the T_c of the trilayer decreases quickly and monotonically from the T_c of a Nb/Cu bilayer until the thickness of the CoFe layer reaches about 2 nm, and then increases slightly to approach a limiting value around $d_{CoFe}\approx 6$ nm, resulting in shallow dip feature. The position and depth of the dip are $d_{Cu}\approx 2$ nm and 60 mK, respectively. This feature is similar in quality to the feature of the Nb/CoFe bilayer[22] and can be understood through the FFLO framework,[23,24] too. In Nb/Cu/CoFe trilayers, the plateau shown in the Nb/Au/CoFe trilayer[25] due to lack of good wetting does not exist, again consistent with a smoother morphology and a lower interface resistance.

By calculating the T_c of a Nb/Cu/CoFe(d_{CoFe}) trilayer using the same method as above, we obtained the same fitting result as for a Nb/Cu(d_{Cu})/CoFe trilayer, except for a slightly different $\gamma_b^{NF}=0.46$. We can clearly see that this fit-

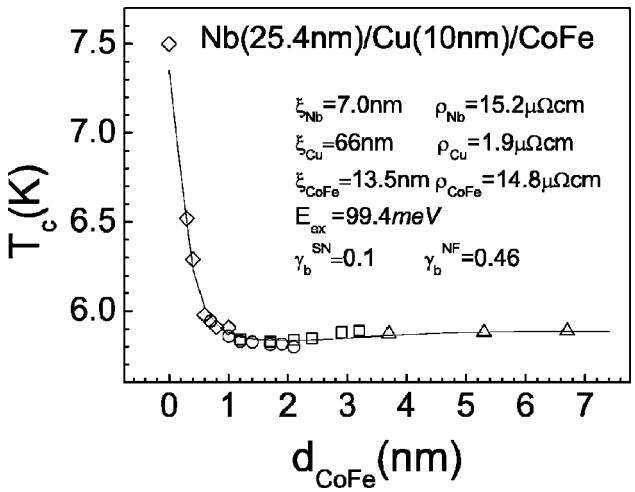

FIG. 5. T_c behavior of Nb(25.4 nm)/Cu(10 nm)/CoFe trilayers with respect to CoFe thickness. The same symbol type represents the samples from the same holder. The solid line is a calculation result explained in the text.

TABLE I. Summary of the materials parameters of the three materials used in our trilayers.

Material	ξ (nm)	ρ ($\mu\Omega$ cm)	E_{ex} (meV)	γ_b^{SN}	γ_b^{NF}	RMS roughness (nm) S	S/N	$S/N/F$
Nb	7.0	15.2				0.17		
				0.1				
Cu	66	1.9					0.19	
					0.44–0.46			
CoFe	13.5	14.8	99.4					0.19

ting result is in good agreement with our data. This shows that the parameters we deduced from the fitting of our data by Usadel formalism are consistent throughout our experiments.

In summary, we have fabricated Nb/Cu/CoFe trilayers in which we are able to control the coupling between the superconductivity and the ferromagnetism by inserting a normal metal layer, Cu. We have presented the T_c behavior in S/N bilayer and S/N/F trilayers. We analyzed these by using the quantitative method based on Usadel formalism and obtained a good agreement between our data and theoretical prediction except for the thin Cu thickness range in the Nb/Cu(d_{Cu})/CoFe trilayer. The length scale for the initial T_c increase, about 2.2 nm, for the Nb/Cu(d_{Cu})/CoFe trilayer seems to be the distance necessary for the effect of both interfaces to come into play fully independently. However, the T_c oscillation behavior as a function of d_{N} in the case of Nb/Au/CoFe was not observed in the case of Nb/Cu/CoFe. In the Nb/Cu/CoFe(d_{CoFe}) trilayer, we observed a similar dip structure to that found in the Nb/CoFe bilayer, consistent with the parameters obtained from Usadel formalism (Table I).

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*Author to whom correspondence should be addressed. Electronic address: kchar@phya.snu.ac.kr

†Current address: Department of Physics, University of Toronto, Toronto, Ontario M5S 1A7, Canada.

[1] H. Meissner, Phys. Rev. **117**, 672 (1960).
[2] J. J. Hauser, H. C. Theuerer, and N. R. Werthamer, Phys. Rev. **136**, A637 (1964).
[3] P. Hilsch, Z. Phys. **167**, 511 (1962).
[4] P. G. de Gennes and E. Guyon, Phys. Lett. **3**, 168 (1963).
[5] P. G. de Gennes, Rev. Mod. Phys. **36**, 225 (1964).
[6] N. R. Werthamer, Phys. Rev. **132**, 2440 (1963).
[7] A. I. Buzdin and M. Yu. Kupriyanov, JETP Lett. **52**, 487 (1990).
[8] Z. Radovic, M. Ledvij, L. Dobrosavljevic-Grujic, A. I. Buzdin, and J. R. Clem, Phys. Rev. B **44**, 759 (1991).
[9] C. Strunk, C. Surgers, U. Paschen, and H. v. Lohneysen, Phys. Rev. B **49**, 4053 (1994).
[10] L. V. Mercaldo, C. Attanasio, C. Coccorese, L. Maritato, S. L. Prischepa, and M. Salvato, Phys. Rev. B **53**, 14040 (1996).
[11] J. E. Mattson, R. M. Osgood III, C. D. Potter, C. H. Sowers, and S. D. Bader, J. Vac. Sci. Technol. A **15**, 1774 (1997).
[12] J. Aarts, J. M. E. Geers, E. Brück, A. A. Golubov, and R. Coehoorn, Phys. Rev. B **56**, 2779 (1997).
[13] E. A. Demler, G. B. Arnold, and M. R. Beasley, Phys. Rev. B **55**, 15174 (1997).
[14] L. R. Tagirov, Physica C **307**, 145 (1998).
[15] M. Velez, M. C. Cyrille, S. Kim, J. L. Vicent, and Ivan K. Schuller, Phys. Rev. B **59**, 14659 (1999).
[16] 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).
[17] O. Bourgeois and R. C. Dynes, Phys. Rev. B **65**, 144503 (2002).
[18] 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).
[19] Ya. V. Fominov, N. M. Chtchelkatchev, and A. A. Golubov, Phys. Rev. B **66**, 014507 (2002).
[20] V. V. Ryazanov, V. A. Oboznov, A. S. Prokof'ef, and S. V. Dubonos, JETP Lett. **77**, 39 (2003).
[21] A. S. Sidorenko, V. I. Zdravkov, A. A. Prepelitsa, C. Helbig, Y. Luo, S. Gsell, M. Schreck, S. Klimm, S. Horn, L. R. Tagirov, and R. Tidecks, Ann. Phys. **12**, 37 (2003).
[22] Jinho Kim, Jun Hyung Kwon, K. Char, Hyeonjin Doh, and Han-Yong Choi, Phys. Rev. B **72**, 014518 (2005).
[23] P. Fulde and R. A. Ferrell, Phys. Rev. **135**, A550 (1964).
[24] A. Larkin and Y. Ovchinnikov, Sov. Phys. JETP **20**, 762 (1965).
[25] Jinho Kim, Yong-Joo Doh, K. Char, Hyeonjin Doh, and Han-Yong Choi, Phys. Rev. B **71**, 214519 (2005).
[26] H. Yamazaki, N. Shannon, and H. Takagi, Phys. Rev. B **73**, 094507 (2006).
[27] K. D. Usadel, Phys. Rev. Lett. **25**, 507 (1970).
[28] Nayoung Lee, Han-Yong Choi, Hyeonjin Doh, K. Char, and Hyun-Woo Lee, preprint.
[29] H. W. Weber, E. Seidl, C. Laa, E. Schachinger, M. Prohammer, A. Junod, and D. Eckert, Phys. Rev. B **44**, 7585 (1991).