Flux Pinning in Multifilamentary Superconducting Wires with Ferromagnetic Artificial Pinning Centers

J.-Q. Wang, N. D. Rizzo, and D. E. Prober
Dept. of Applied Physics and Physics, Yale University, New Haven, CT 06520

L. R. Motowidlo and B. A. Zeitlin
IGC - AS, Waterbury, CT 06704

***Abstract*—We demonstrate that ferromagnets are effective artificial pinning centers for the enhancement of critical current (J_c) in multifilamentary superconducting wires. We have analyzed theoretically the proximity effect due to the FM pins near the final size of several nanometers and determined that one should achieve a large pinning force by such centers at these sizes. There is also additional pinning strength resulting from the interaction between ferromagnetic moments of the pins and the magnetic field gradient of the fluxon lattice. The measured results of J_c, T_c and H_{c2} are analyzed, and compared with our model analysis.**

I. Introduction

Multifilamentary superconducting wires are used in many device applications, such as (MRI) magnetic resonance imaging. With increased critical currents (J_c), the range of application will be extended, and may include electric power generators with reduced weight and cost. One novel approach to increase J_c is to introduce artificial pinning centers (APC) with a flexible choice of pin material, size and density [1]-[5]. The pin material has a strong effect on the superconducting properties and influences the design of APC wires. Among various ductile materials, ferromagnetic (FM) metals such as Ni and Fe, deserve special attention because of their very strong suppression of superconductivity. We have recently reported [6] the results of J_c for Ni and Fe APC wires that we have fabricated to demonstrate the effectiveness of FM pin materials. The J_c's are higher, in specific field ranges, than commercial MRI wires, and further improvement should be possible by varying the pin design.

In this report we investigate flux pinning by FM pins for enhanced J_c in NbTi wires, and explore the design parameters and practical limitations for improved APC wires. First, the superconducting order-parameter is calculated in the vicinity of an idealized FM pin. The elementary and bulk pinning forces are estimated from the derived order-parameter. We compare these results with our measured values of J_c for several FM APC wires. We conclude that core pinning is the dominant mechanism in our APC wires. We further investigate another possible pinning mechanism, the magnetic interaction, and estimate its pinning strength. Finally, proximity-effect-induced T_c and H_{c2} depression is analyzed and compared with the corresponding measured values for some FM APC wires.

Manuscript received August 27, 1996.

II. Flux Pinning By FM Pins

A FM material is an effective Cooper-pair breaker, since its strong exchange interaction tends to act against the anti-parallel coupled spins of the pair. In proximity to a superconductor, it forces the order-parameter to almost zero at the interface, with a very small amount penetrating into the FM region. The penetration length in the ferromagnet is very short, much smaller than the coherence length of the superconductor (ξ); e.g., 0.9nm for Ni and 0.6nm for Fe [7], whereas $\xi = 4 \sim 5$nm in NbTi.

The FM APC wires we have fabricated contain a small volume percent of FM pins to avoid proximity-effect induced degradation. For all the APC wires we made, at the optimal wire sizes (i.e., for maximum J_c), the average pin spacing (D_{pp}) has reached about 30nm, comparable to the flux lattice spacing (a_B) at 5T ($\sim$ 22nm). At this pin spacing, the FM pin diameter is about 4nm. The question is, in this two dimensional (2-D) case, how a circular pin of such size quantitatively changes the order-parameter profile near the pin, which in turn determines the free energy profile and hence the pinning force. The order-parameter in a system in the presence of a ferromagnet is complex due to the proximity of the ferromagnet. We use a phenomenological Ginzberg-Landau (G-L) theory based on generalizing a quasiclassical treatment of superconducting/ferromagnet superlattice in the dirty limit. [8] In this treatment, the pair amplitude function $F_M(\vec{r})$ in the ferromagnet satisfies the following linearized equation

$$\nabla^2 F_M(\vec{r}) = i/{\xi_M}^2 F_M(\vec{r}), \tag{1}$$

where

$$\xi_M = (\hbar D_M/2I)^{1/2} \tag{2}$$

is the characteristic decay distance of F_M in the ferromagnet. In the above equation, the coefficient D_M is the diffusion constant for the ferromagnet, I the exchange interaction strength, and $\vec{r}$ the 2-D spatial vector. We identify $F_M(\vec{r})$ with the G-L order-parameter, assuming that it also satisfies (1). Higher than linear-order terms are not

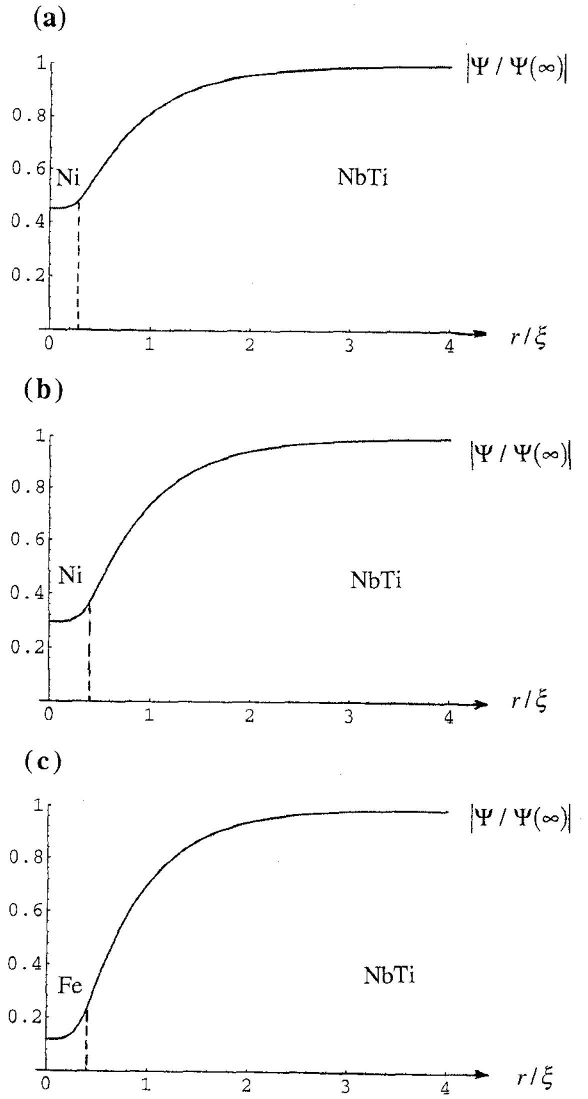

Fig. 1. Superconducting order-parameter near a circular ferromagnetic pin. (a) A Ni pin of diameter d_p = 3nm. (b) A Ni pin of d_p = 4nm. (c) A Fe pin of d_p = 4nm.

included since $F_M(\vec{r})$ is usually small in ferromagnet. The solution of F_M is simply the zeroth order Bessel function

$$F_M(\vec{r}) = J_0(|\vec{r}|e^{i\pi/4}/\xi_M), \tag{3}$$

with a complex argument. In the superconductor, the order-parameter satisfies the G-L equation, including the $|F|^2F$ term.

In making FM APC wires, we have chosen high purity Ni and Fe for the pin material. Therefore, the electron mean free path in the pin ($l_{\rm FM}$) is likely limited by scattering at the pin boundary. Assuming the same electron density of states in the two phases, the Zaitsev boundary conditions [9] are expected to be satisfied. Hence, at the pin boundary , we have

$$\frac{\nabla F}{F} = \eta \frac{\nabla F_M}{F_M}, \tag{4}$$

with $\eta = 1$. Generalizing DeGennes dirty-limit boundary conditions [10], we have for (4) $\eta = (l_{\rm FM}/l)^{1/2}$, where $l \approx 0.5nm$ is the mean free path in NbTi. The estimated value of η is 3. In fact, due to the small pin size, it matters very little to the pinning force as to the detailed shape of the order-parameter in the pin.

With these constraints, $F(r)$ in the superconductor is solved exactly by numerical methods using Mathematica. In Fig. 1a, 1b and 1c, we show the order-parameter profile, respectively, around a Ni pin of 3nm, 4nm, and a Fe pin of 4nm in diameter. For the Fe pin, we observe that a very small amount of the order-parameter is present in the pin: $F(\vec{r})$ is very small at the pin interface and it extrapolates to zero at $r \approx 0$. It recovers 90% of the full bulk value within a coherence length ξ. To estimate the elementary pinning force from the calculated $F(\vec{r})$ profile, the ferromagnetic pin can be approximated by a void of diameter $2\xi + d_p$, where d_p is the actual pin diameter. It is a standard result that the pinning force of a void of volume V_{pin} is

$$f_p \approx \frac{H_{c2}^2}{(2\kappa^2)(8\pi)} \frac{V_{pin}}{\xi}, \tag{5}$$

where $V_{pin} = \pi\xi^2(2\xi + d_p)$ when the fluxon is perpendicular to the pin. We have deduced the values of κ and ξ from H_{c2} and T_c measurements for our FM-APC wires and obtain $\kappa \approx 40$ and $\xi \approx 4.5$nm. From these results and H_{c2} = 11T, the estimated pinning force is $f_p \approx 2.7 \times 10^{-7}$dyn.

The field dependence of f_p is $(1 - B/B_{c2})$, with B_{c2} being the upper critical field of the composite wire. Assuming a flux-lattice matching with the pins for the optimal wires made (i.e., $a_B = D_{pp}$), the pin density is $n_p = 1/({D_{pp}}^3)$. We estimate a value for the bulk pinning force F_p = 60GN/m^3, which is higher than our observed value 13 GN/m^3 at 3T. There are several reasons for this difference: 1. The above estimate is accurate only within a numerical factor, probably of order of two. 2. The estimate assumes complete order-parameter suppression at the pin boundary, which is not the case, especially for the APC wires with small pin percent near final sizes. 3. The pins may be distorted after drawing, reducing the effectiveness of pinning. 4. There is a loss of ferromagnetism at the actual pin sizes for our wires, which further reduces the effective pin size. For a 2% Ni wire, only 1% pin material remains ferromagnetic at the optimal pin diameter (4nm). The FM magnetization density M should be the bulk value even for FM wires with the small pin diameters, since the M remains unchanged in small FM spheres down to 2nm in diameter [11]. We conclude that the FM pins have a smaller effective size than the design value. It appears that the remaining pin material mechanically mixes with surrounding materials, forming a nonmagnetic alloy. Thus, the pins have a smaller effective pin size, around 3nm instead of 4nm. As shown in Fig. 1a, there is substantial order-parameter present in the Ni pin for a 3nm effective diameter.

III. Pinning By Magnetic Interaction

There have been studies of how the magnetic energy of the flux lattice influences the flux pinning properties. In a system with ferromagnetic pinning centers, there are two aspects of magnetic pinning. One is the usual magnetic pinning energy which exists in all systems with heterogeneous phases. It is associated with magnetic field redistribution of the vortex when it enters into one phase from the other. The change in magnetic field profile results in a variation in magnetic energy, and a pinning force. One example is the surface barrier pinning at an interface of a superconductor and a non-superconducting metal [12], such as Cu. In the APC wires with a submicron superconducting filament size, embedded in Cu matrix, this surface barrier pinning can be substantial at low field, as observed both by us [6] and others [13].

The magnetic pinning by APC structures, which are non-ferromagnetic, is rather weak compared with the core pinning. A theoretical study by Tachiki *et al* indicates that magnetic pinning is only 15% of the core pinning discussed in the above section [14]. The reason for the weak magnetic pinning in APC wires is the relatively long penetration depth ($\lambda = 0.14\mu$m) compared with the small pinning centers. Thus, the nanometer sized APC structure does not significantly alter the vortex structure, and the associated fields. In this paper, we are concerned with another aspect of magnetic pinning which is unique to magnetic pinning centers. This is the magnetic interaction between the magnetic moment of the FM pins and the magnetic field of the flux lattice. Earlier studies have investigated this question, in searching for novel pinning mechanisms [15, 16]. The periodic vortex structure in the flux lattice produces a periodic variation of magnetic field distribution. At the centers of the fluxon cores the field is the highest, whereas at the points furthest from the cores the field is minimum. The variation has a small magnitude in the intermediate and high field ranges in type II superconductors, such as NbTi. However, some FM pins, such as Fe, have a strong magnetic moment. With an aligned magnetization in an applied field, the magnetic energy of the interaction between the FM pins and magnetic field distribution is the lowest when a fluxon is on the pins and the reverse is true when it is in between the FM pins. In other words, the fluxons prefer to stay on the FM pins because of this magnetic interaction, resulting in a pinning.

To estimate the strength of the pinning force, the spatial variation of the magnetic field is approximated as sinusoidal. The magnitude of the variation (ΔH) can be derived from the calculated induced magnetization of a type II superconductor in applied magnetic field (H) [17]. In an intermediate field, $\xi^2 \ll \frac{\Phi_0}{H} \ll \lambda^2$, we have

$$\Delta H = (2H_{c2}/\kappa^2)ln[\alpha/\xi(H/\Phi_0)^{1/2}], \qquad (6)$$

where α is a constant of the order of unity. In NbTi, we have for H=5T, $\Delta H = 0.02$T. Assuming perfect flux lattice and pin array matching, the pinning force for an optimal 2% Fe APC wire (with nearest neighbor pin spacing 30nm) is estimated to be 23GN/m^3. This value is comparable in strength to the estimated core pinning, hence should contribute significantly to the pinning. If the lattice match is not perfect, F_p will be smaller. This is the case for our wires.

IV. Proximity-Effect Induced T_c Reduction

In designing APC superconducting wires and in evaluating their performance, a crucial consideration is the influence of APC structure on the critical temperatures and fields. These parameters directly relate to the critical current and they are affected by such sample parameters as pin material, size and separation. Severe T_c (and H_{c2}) depression results in lower J_c's. Given the lengthy process to fabricate the APC structure starting from macroscopic sizes, it is desirable to be able to predict the expected T_c and H_{c2}. This allows one to avoid designs which may result in degraded performance.

T_c and H_{c2} in heterogeneous superconducting systems are strongly influenced by the microstructure because of proximity effects. When the size scale of the microstructure variation becomes comparable to the coherence length, the superconducting order-parameter is modified by the presence of the foreign phase in the system. In APC wires, the modified order-parameter has to have the same periodicity as the APC structure. This requirement sets a constraint on the critical temperature of the system. For FM APC's the solution to the modification of T_c is relatively simple, since in the limit of thick pins the order-parameter is forced to become zero at the pins. In the optimal wires, however, the order-parameter likely retains some finite value at the pins.

The situation of complete Cooper-pair destruction in the FM pins provides an interesting test case for modelling. Similar to the analysis of one-dimensional (1-D) superconductor/ferromagnet multilayers [18], we found a solution of the linearized Ginzberg-Landau equation in the 2-D case which satisfies both the periodicity and the boundary conditions at the pins. Approximating the temperature dependence of the coherence length as $\xi(T) = \xi/[1-(T/T_{c0})^2]^{1/2}$ near T_c, the above conditions require that the following relation is satisfied for T_c

$$T_c = T_{c0}\left[1 - \frac{1}{2}\left(\frac{\pi\xi}{D^e_{pp}}\right)^2\right], \qquad (7)$$

where T_{c0} is the critical temperature for the bulk superconductor in the absence of the proximity effect. Thus, the T_c suppression is found to be $\Delta T_c/T_c = \frac{1}{2}\left(\pi\xi/D^e_{pp}\right)^2$, with D^e_{pp} being the effective pin spacing. The suppression of T_c correlates with that of H_{c2} as $\Delta H_{c2}(0)/H_{c2}(0) = \Delta T_c/T_c$. $H_{c2}(0)$ is the upper critical field at $T = 0$. To find this relation at the operating temperature, we use the measured $H_{c2} - T_c$ in NbTi [19], and find that, at $T = 4.2K$ ($T_c = 9K$), $\Delta H_{c2}(T)/H_{c2}(T) \approx 1.6\Delta T_c/T_c$.

These results can be tested by an APC wire made with a larger Fe pin percent. We made a 4% Fe wire. To prevent loss of ferromagnetism at the final sizes, we used a 9% Cu sleeve around the Fe pin. We found that at the final size, there is about 3% Fe remaining ferromagnetic. There is a substantial H_{c2} and T_c suppression in this wire. From bulk pinning force curve we derive an H_{c2} of 8.2T. The T_c is found to be 7.6K by magnetic measurement. The above model gives 8.8T for H_{c2} and T_c = 7.9K near the optimal size. Apparently, the additional Cu is giving further H_{c2} and T_c reduction. Extending this model analysis to the APC wire with 2% Ni in a 3% Cu sleeve, we predict a T_c of 8.3K and an H_{c2} of 9.8T, compared with our measured values of 8.6K and 9.6T.

An earlier study has analyzed proximity effect and flux pinning by $\alpha -$ Ti ribbons in the NbTi superconductor, and related the H_{c2} and T_c to the maximum local free energy of the order-parameter [20]. If we use that analysis for the 4% Fe wire we would predict a much smaller T_c suppression (1%) than observed. We have also tested the 1-D situation, by a numerical method. Using the exact solution for T_c suppression, and correlating it with the maximum order-parameter we find that $\Delta T_c/T_c \approx (\delta F/F)^{0.5}$, unlike the relation $\Delta T_c/T_c \approx (\delta F/F)^2$ proposed [20]. The quantity $\delta F/F$ is the fractional change of the order parameter at the center of a superconducting layer.

V. Conclusions

We have analyzed pinning mechanisms and proximity effects in FM APC superconducting wires, and compared the predictions with the experimental results. Our study has demonstrated that ferromagnets are strong pinning centers by creating a void in the superconducting order-parameter. For a strong ferromagnet, even with a small pin size, the size of the void can be comparable to the fluxon core. We find that core pinning is the dominant mechanism responsible for the observed J_c. We have analyzed the T_c and H_{c2} of APC structure and find a stronger constraint on these parameters than earlier realized. This can be useful for future APC design involving FM pinning centers.

Acknowledgment

This research was supported by IGC and CT Dept. of Econ. Development. We wish to thank N. Ivin for discussions and J. Budnick and G. Luke for SQUID measurements at Univ. of Conn. and Columbia Univ., respectively.

References

[1] L. R. Motowidlo, B. A. Zeitlin, M. S. Walker, and P. Haldar, Appl. Phys. Lett. , vol. 61, pp. 991, 1992; H. C. Kanithi, P. Valaris, L. R. Motowidlo, B. A. Zeitlin and R. Scanlon, Adv. Cryog. Eng. , vol. 38B, pp. 675, 1992.

[2] L. R. Motowidlo, B. A. Zeitlin, M. S. Walker, P. Haldar, J. D. McCambridge, N. D. Rizzo, X. S. Ling, and D. E. Prober, IEEE Trans. Appl. Supercond., vol. 3, pp. 1366, 1993.

[3] K. Matsumoto, H. Takewaki, Y. Tanaka, O. Miura, K. Yamafuji, K. Funaki, M. Iwakuma, T. Matsushita, Appl. Phys. Lett., vol. 64, pp. 115 , 1994.

[4] L. D. Cooley, P. J. Lee, and D. C. Larbalestier, Appl. Phys. Lett., col. 64, pp. 1298, 1994.

[5] M. K. Rudziak, J. M. Seuntjens, C. V. Rebaud, T. Wong, and J. Wong, IEEE Trans. Appl. Supercond., vol. 5, pp. 1709, 1995.

[6] J.-Q. Wang, N. D. Rizzo, J. D. McCambridge, D. E. Prober, L. R. Motowidlo, and B. A. Zeitlin, Adv. Cryog. Eng., vol. 42, 1995; N. D. Rizzo, J.-Q. Wang, D. E. Prober, L. R. Motowidlo, and B. A. Zeitlin, to appear in Appl. Phys. Lett. in Oct. 1996.

[7] J. J. Hauser, H. C. Theuerer, and N. R. Werthamer, Phys. Rev., vol. 142, pp.118, 1966.

[8] Z. Radović, L. Dobrosavljević-Grujić, A. I. Buzdin, J. R. Clem, Phys. Rev. B, vol. 38, pp. 2388, 1988; Z. Radović, M. Ledvij, L. Dobrosavljević-Grujić, A. I. Buzdin, J. R. Clem, Phys. Rev. B, vol. 44, pp.759, 1991.

[9] R. O. Zaitsev, Sov. Phys. JETP, vol. 23, pp. 702, 1966.

[10] P. G. DeGennes, Rev. Mod. Phys., vol. 36, pp. 225, 1964.

[11] J.-Q. Wang and G. Xiao, Phys. Rev. B, vol. 49, pp. 3982, 1994.

[12] C. P. Bean and J. D. Livingston, Phys. Rev. Lett., vol. 12, pp. 14, 1964.

[13] L. D. Cooley and D. C. Larbalestier, Proc. 8th US-Japan Workshop, Univ. of Wisconsin, Madison, 1993.

[14] M. Tachiki, S. Takahashi, and K. Sunaga, Phys. Rev. B, vol. 47, pp. 6095, 1993.

[15] T. H. Alden and J. D. Livingston, J. Appl. Phys., vol. 37, p. 3551, 1966.

[16] S. H. Autler, J. Low Temp. Phys., vol. 9, pp. 241, 1972.

[17] D. Saint-James, E. J. Thomas, G. Sarma, *Type II Superconductivity,* New York: Pergamon, 1966, pp. 69.

[18] B. Y. Jin and J. B. Ketterson, Adv. Phys. vol. 38, pp. 189, 1989.

[19] Y. Shapira and L. J. Neuringer, Phys. Rev., vol 140A, pp. 1638, 1965.

[20] T. Matsushita, S. Otabe, T. Matsuno, Adv. Cryog. Eng., vol. 36, pp. 263, 1990.