Superconducting Properties and Critical Current Density of Nb-Ti/Ti Multilayers

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Abstract—Multilayers of Nb47Ti superconductor (S) and titanium pins (N) have been made with a critical temperature T_c and an upper critical field H_{c2} approaching bulk values. There is no proximity effect suppression of T_c and H_{c2} for layer thickness $d_S = d_N = 10$ nm, in contrast to the strong suppression for comparable multilayers made with Cu, Cu-alloy, or Nb pins. This may be because the proximity length of the Ti layers, ~10 nm, is less than or equal to their thickness. The critical current density J_c exhibits multiple peaks in $J_c(H)$, which suggests that matching effects contribute to the overall pinning. J_c for 20 nm bilayers is comparable to that of Nb47Ti tapes, which have a higher number density of pins but a lower pin volume fraction. The weak proximity coupling suggests that much higher J_c can be obtained if smaller bilayer periods can be made with good adhesion. The results are compared to other Nb-Ti/Ti multilayer experiments, and stability limitations are also discussed.

I. INTRODUCTION

Niobium-titanium alloy composites attain strong flux pinning and high critical current density J_c when a nano-structure of the superconducting and non-superconducting phases is made. In conventional wires, non-superconducting α-Ti precipitates are the pins and have 1-4 nm thickness and 4-10 nm separation when flux pinning is optimum [1]. In artificial pinning-center (APC) composites with Nb pins, the optimum nanostructure is somewhat less refined, the pins having 10-15 nm thickness and 10-30 nm separation [2]. Both conventional and APC composites also exhibit proximity effects when the optimum nanostructure is obtained, characterized by changes in the critical temperature T_c and the upper critical field H_{c2} [2]-[4]. The differences in the optimum flux-pinning states and the connection to the proximity effect were explained by Cooley et al. [2], [5] in terms of a magnetic pinning mechanism, where the proximity length ξ_N is thought to be the parameter that determines the optimum nanostructural state.

In both conventional and APC composites, the pins and the superconductor take the form of ribbon-like regions that are folded and curled together [1]-[3]. Thus, multilayers of Nb-Ti and various pin materials have been used to model the pinning force and separate out the effects of orientation of the pin to the field [6]-[8]. Multilayers also give the flexibility to vary the pin material without concern for ductility and other fabrication issues. Multilayers of Nb47Ti with Cu pin layers exhibited $J_c \sim 4$ kA/mm^2 at 5 T parallel field and 4.2 K [6],

[9], and they had an upper critical field of 11 T, even though T_c was suppressed. However, efforts to further increase J_c by alloying the pin layer and reducing ξ_N resulted in a greater suppression of superconducting properties [10]. Multilayers with Nb pin layers have been extensively explored because they exhibit no suppression of T_c and interesting properties of H_{c2} [7], [11]. In this case J_c is ~10 kA/mm^2 [7], although the field dependence $J_c(H)$ is not monotonic and H_{c2} is suppressed. Multilayers with Ti pins can achieve $J_c > 20$ kA/mm^2 at 5 T parallel field and 4.2 K [7]. This is a factor of 4 higher than the J_c of a Nb-Ti tape with α-Ti precipitates aligned to within ~10° of the tape face [12]. This result is intriguing because the bilayer period $\Lambda = d_S + d_N$ of the multilayer in [7] is somewhat larger than the average precipitate spacing in optimized flattened conventional composites [12], ~20 vs. ~5-10 nm. Further increase of J_c may thus come if the pin thickness d_N and the superconductor thickness d_S can be reduced to a few nanometers. However, the presence of thick Nb buffer layers masked the superconducting properties of that multilayer, and there is little other characterization to explain this singular result.

The superconducting properties and critical current density of multilayers of Nb47Ti superconductor (S) and Ti pin (N) layers are examined in this paper. Superconducting properties close to those of bulk specimens were obtained for thin films of Nb47Ti, as is discussed more completely in another paper [13]. These properties are maintained in the multilayers by using the same deposition parameters as for the films. A surprising result is $T_c = 8.6$ K and $\mu_0H_{c2} = 11.4$ T, even for $d_S = d_N = 10$ nm (10/10), about twice the coherence length $\xi = (\phi_0/2\pi\mu_0H_{c2})^{1/2} \approx 5$ nm. This suggests that the proximity effect is weak, perhaps because ξ_N is ~10 nm, as deduced from the resistivity. The superconducting layers then appear to behave independently when $d_N = 15$ nm $> \xi_N$, where peaks in $J_c(H)$ suggest the penetration of flux-line rows into the S layer when d_S or Λ is a multiple of the flux-line spacing $a_0 \approx (\phi_0 / \mu_0H)^{1/2}$. Similar phenomena are found for the 10/10 multilayer, where J_c has 2 plateaus and drops from ~8 kA/mm^2 at 4 T to ~5 kA/mm^2 at 5 T and ~2 kA/mm^2 at 6 T parallel field, when $a_0 \approx \Lambda$. The J_c values for the 10/10 multilayer are comparable to the tape results in [12], but for about one-half the pin number density.

Because of the weak proximity effect, further increase in J_c should thus be possible if 1-5 nm layers can be made and the associated adhesion problems solved [13]. Transmission electron microscopy (TEM) shows that the layers are perturbed by columnar grain growth through several layers at this length scale. Although the matching effects and other data suggest that the lack of layer uniformity does not adversely affect flux pinning, the grains may trap gases present during sputtering, which adds stress to the multilayer. Poor adhesion has so far prevented exploration of smaller bilayer periods. Other limitations, such as poor stability of the multilayer, are also discussed. The present work is also

compared to [7] and several important differences between the experiments are discussed.

II. EXPERIMENT

The multilayers were fabricated by dc sputtering at room temperature on R-plane sapphire, using a technique described previously [6]. A high-homogeneity alloy target was used for the superconductor. The deposition rates were 2.8 and 2.0 nm/s for S and N respectively, at a power density of about 13 W/cm^2. This high deposition rate gave good properties in a 560 nm-thick Nb47Ti film, where T_c = 8.95 K, $\mu_0 H_{c2}$ = 11.3 T at 4.2 K (see Fig. 2), and the resistivity was 72 $\mu\Omega$-cm at T_c. These values are close to the values of bulk Nb47Ti: 9.1 K, 11.1 T, and 65 $\mu\Omega$-cm respectively [3]. The layers are polycrystalline, as observed by TEM. No buffer layer was used; instead, the top and bottom layers are Ti pin layers.

A system of bridges was patterned by standard lithography techniques and wet etching. The bridge widths were measured by light microscopy using a calibrated length standard, and the effects of undercutting during the etching process were characterized by scanning electron microscopy. The multilayer thickness was determined by profilometer measurements. The bridge width used for the measurements was 35-60 μm, and the multilayer thickness was 1 μm. The voltage taps were 1 cm apart. Wires were then connected to the bridges by either using spring-loaded pins or by bonding them directly with indium. All critical current measurements using an apparatus described previously [6]. Measurements at 4.2 K were in liquid helium, and flowing He gas and a temperature controller were used at higher temperature. An electric field criterion of 0.1 μV/cm was used to determine the critical current. The value of J_c was then determined by dividing the critical current by the cross-section of the bridge.

The superconducting properties of 2 of these multilayers are given focus in this paper, along with the Nb47Ti film described above. One had d_S = 45 nm and d_N = 15 nm (45/15), and the other had d_S = 10 nm and d_N = 10 nm (10/10). The 45/15 film was studied because d_S was much larger than ξ but d_N was thought to be less than ξ_N [5], while the 10/10 film was the smallest bilayer period which could be made with good adhesion and was thought to be fully proximity coupled. The critical temperature of these multilayers is shown in Table I. The value of T_c was taken as the temperature at which resistance was zero for a constant current of 1 μA. The transitions were less than 0.1 K wide. The resistivity of the Ti layer ρ_N and the proximity length are also given in Table I. These values were found by applying the parallel resistance law to the measured resistance, assuming that the resistivity of the S layers is equal to that of the Nb47Ti thin film. In previous work [5] the value of ξ_N was determined by using the resistivity of a 0.23 mm diameter Ti wire, ρ_0 = 5.6 μΩ-cm. Since ξ_N is proportional to
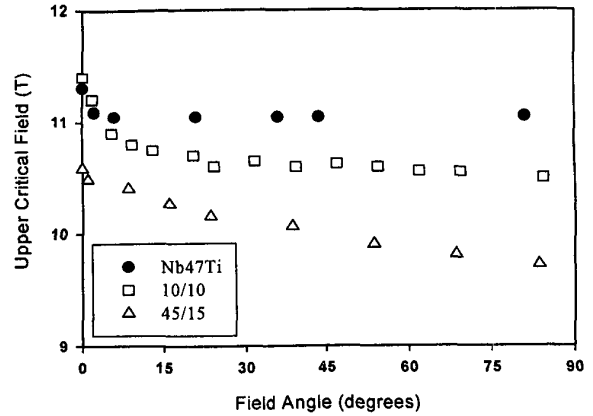

Fig. 1. Upper critical field as a function of the field angle for the Nb47Ti film and the multilayers. 0 degrees represents parallel field.

$(1/\rho_N)^{1/2}$, the value in [5], 32 nm, was multiplied by $(\rho_0/\rho_N)^{1/2}$ to obtain the values in Table I.

The upper critical field as a function of field angle θ at 4.2 K is shown in Fig. 1. The field at which the resistance was 1% of the normal-state resistance R determined these data. A constant current of 1 μA was applied while changing the field at fixed angle. The resistive transitions were very broad for the Nb47Ti film and the multilayers, with approximately 2 T between 10% and 90% of R. Thus there is evidence of superconductivity even at ~12 T, 4.2 K, which may be due to the increased resistivity of the Nb47Ti layers. H_{c2} (θ) has a small upturn and cusp near parallel field for both the 10/10 multilayer and the Nb47Ti film. Above ~15° the angular dependence is weak. Both of these samples exhibit high values of $\mu_0 H_{c2}$, reaching 11.3 T in parallel field and 10.8-11.0 T in perpendicular field. A moderate angular dependence is seen for the 45/15 multilayer, with somewhat lower values for the upper critical field at all angles than for the thin film and the 10/10 multilayer.

The critical current density as a function of parallel field at 4.2 K is shown in Fig. 2. The black symbols show J_c data for 45/15 and 10/10. It can be seen that these data do not decrease monotonically with field as for the Nb47Ti film, shown by the dot-dash line, but have a series of peaks. This is more pronounced in the bulk pinning-force data, $F_p(H)=\mu_0 H J_c(H)$, shown by the open symbols. The peaks are especially prominent in the 45/15 data. Smoother plateaus are observed for the 10/10 multilayer. The data for 10/10 is limited to ~8 kA/mm^2 because the sample quenched for the smallest bridge width tested, 35 μm. Apparently the value of J_c increases between ~2 and ~4 T because stable current-voltage curves were acquired at 1.8, 2, and 2.25 T. There is a sharp drop in the J_c of the 10/10 multilayer in the mid-field regime, from ~8 kA/mm^2 at 4 T, to ~5 kA/mm^2 at 5 T and ~2 kA/mm^2 at 6 T.

III. DISCUSSION

The body of literature about Nb-Ti thin films and multilayers suggests that it is difficult to obtain superconducting properties comparable to those of bulk alloys by

TABLE I

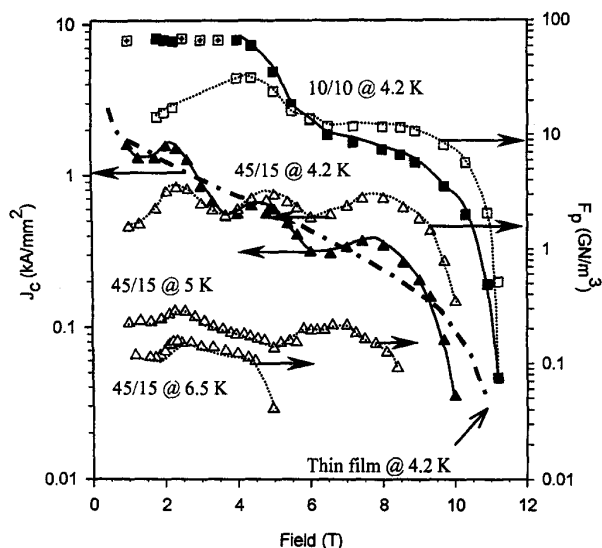| Multilayer	T_c (K)	ρ_N (μΩ-cm)	ξ_N (nm)
45/15	8.45	56	10
10/10	8.6	51	11

Fig. 2. Critical current density and bulk pinning force as a function of the applied parallel field. Critical current density for the 45/15 and the 10/10 multilayers are represented by the black triangles and black squares respectively. The white squares with crosses denote values at which a quench occurred. For comparison, the critical current density of the 560 nm Nb47Ti film is given by the dash-dotted line. Open symbols denote the bulk pinning force, as labeled near the curve.

sputtering. Generally the highest T_c values are 8.6 K or less when the superconductor is Nb47Ti [6], [7], [14], and the resistivity is 75 $\mu\Omega$-cm or higher [6], [14]. In a companion paper [15], it is shown that this may be due to the complicated nature of the sputtering process.

Nonetheless, the Nb47Ti film has better superconducting properties than reported previously. Using the same deposition parameters, good properties in multilayers are also obtained. The variation of the critical temperature between the thin film and the multilayers is less than the variation of T_c for thin films made at the same power setting over the lifetime of the target [15]. This suggests that there is no clear evidence for suppression of T_c by the proximity effect. By contrast, T_c was strongly suppressed for Nb47Ti multilayers with pure Cu [6] or Cu-alloy [10] pins. In that case 45/15 multilayers had a T_c between 8 and 8.2 K, and 10/10 multilayers had T_c < 6.5 K. The apparent lack of proximity effect suppression is also indicated by the upper critical field data. For the most part, $H_{c2}(\theta)$ is not strong. The small cusp in parallel field for the 10/10 multilayer indicates a somewhat enhanced H_{c2} value of 11.4 T, which could be due to a dimensional crossover of the S layers [15] or surface superconductivity. These data are in contrast to data for Cu-alloy [6], [10] and Nb [11] pin layers, which exhibit a change by a 1-4 T in the value of H_{c2} between parallel and perpendicular field.

The high resistivity of the Ti layers, ~50 $\mu\Omega$-cm, may be beneficial. The proximity length of pure Nb, pure Ti, and Nb-alloys were compared in [5]. This showed that pure Ti had a similar value of ξ_N as Nb10wt.%W. However, in the

present case the resistivity is much higher than the value used in [5], resulting in an even smaller value of ξ_N, about 10 nm. More recent work [10] argues that the proximity length of Cu30at.%Ni, a strong alloy of copper, is greater than 20 nm. Thus, pure Ti is very effective at limiting the extent of the superconducting pair function, and the possible deleterious properties of alloying elements are not present.

The behavior of J_c and F_p are interesting. The pinning force for 45/15 shows several peaks at 4.2 K, 5 K, and 6.5 K. At 4.2 K the peaks correspond to $a_0 \approx 30$, 20, and 15 nm at 2.2, 5, and 7.8 T respectively. The 2.2 T peak is also present at 5 K and 6.5 K, but the high-field peaks become broad and move to ~6 T in the 5 K data. The fact that the 2.2 T peak is not dependent on temperature suggests that it is due to a matching effect, $\Lambda = 2a_0$. The other peaks may obey similar configurations for d_S, Λ, and a_0. In this scenario, the first peak corresponds to having 1 flux line row in the center of the S layers and an adjacent row in the N layers; the second to 2 rows in the S layer and 1 in the N layer; and so on. Similar phenomena have been studied in other layered superconductors [16].

The 10/10 multilayer demonstrates much stronger pinning below ~5 T. In this case J_c approaches 8 kA/mm^2 at 4 T and below, and 2 plateaus appear for $J_c(H)$. A steep drop separates the plateaus, occurring when $a_0 = \Lambda$ (~5 T). The plateaus might again be explained by a matching effect; unfortunately, this sample was destroyed after one of the low-field measurements, and data at higher temperature could not be obtained to support this hypothesis. The data suggest that smaller bilayer periods will not only give higher critical current density at low field, but also shift the plateaus to higher flux densities. The critical current density at 5 T, ~5 kA/mm^2, and at 8 T, ~1.5 kA/mm^2, are about the same as the data for a Nb47Ti tape with α-Ti precipitates which were aligned to within ~10° of the tape face. The similarity of these data may reflect a balance between 3 effects: Although the pin number density is somewhat higher in the tape (1 precipitate per 5-10 nm) than in the film (1 pin per 20 nm), the distribution of the precipitate thickness of the tape is broader than the distribution of the pin layer thickness in the multilayer, and the volume fraction of pins in the tape (~20%) is less than that of the multilayer (50%).

The similarity between the tape and the multilayer also suggests that the flux lines are not strongly affected by variations in the layer orientation. To test this hypothesis, we analyzed a 10 nm Nb47Ti / 3 nm Ti multilayer using TEM. This multilayer was made during the same run as the 10/10 multilayer, however it exhibited visible signs of stress during subsequent processing. As shown in Fig. 4, columnar grains grow through several layers and cause a local misorientation of the layers of ~5°. Similar TEM observations were made in [8]. This perturbation is somewhat larger than the angular variation of J_c, which had a cusp at $\theta = 0°$ and a width at half-maximum of ~3°. These data suggest that the flux lines can follow the contours of the pin layers without a significant reduction of the pinning force.

A new limitation of J_c of Nb-Ti/Ti multilayers is poor stability. In contrast to Cu and Nb pin layers, which are good conductors of heat, the thermal conductivity of Ti is about the same as Nb47Ti. This suggests that heat cannot escape

Fig. 4. Transmission electron microscopy image of a 10 nm Nb47Ti / 3 nm Ti multilayer. The sapphire substrate is at the top edge of the photograph. The bright regions are Ti, while the darker gray regions are Nb47Ti . The nearly black regions are grains which strongly diffract the electron beam. The layer structure of the multilayer is evident as horizontal features, while grain boundaries run vertically. Precipitates may also be present, as indicated by the vertical white streaks in the lower left corner.

between the superconducting layers. It was also noted in Fig. 3 that the J_c data was limited to ~8 kA/mm^2 by a quench at low fields. This may be due to a lack of adiabatic stability. By using a standard textbook expression and established values of the specific heat and density of Nb47Ti [17], the critical bridge width for 8 kA/mm^2 current density can be estimated as ~30 μm. In addition, dynamic stability is also poor, where the Stekly number is above 100 [18]. A solution to these stability problems may require the construction of "multifilamentary" bridges, in which 1-10 μm lines are patterned into the multilayer and are subsequently coated with a thick layer of copper.

The critical current density shown in Fig. 3 is not as high as that reported in [7], despite having similar layer dimensions (19/5 in [7]; 10/10 in this paper) and good superconducting properties. However, there are several important differences between the 2 experiments which make the results difficult to compare. In [7], the multilayers contained 10 bilayers and were coated by 50 nm-thick Nb layers on the top and bottom, so it is difficult to assess the critical temperature of the multilayer and how much current might be carried by the Nb. The electric field criterion used to determine the critical current in [7] was 50 μV/cm; here it is 0.1 μV/cm. Given that the current-voltage data obeys a power law with an exponent (n-value) of ~30, the different criteria can give a difference in J_c of ~15%. The bridge width in [7] was 3 μm, which along with the Nb coating would enhance the stability of the multilayer substantially. These differences underscore the variety of different ways that such experiments can be done, and more systematic study of multilayers and their associated physics is needed.

IV. CONCLUSIONS

In conclusion, multilayers of Nb47Ti and Ti were discussed and compared to a 560 nm-thick Nb47Ti film. The film exhibited superconducting properties which were close to those for bulk specimens. Similar parameters were obtained in the multilayers by using the same deposition parameters. The multilayer with 10 nm Nb47Ti and 10 nm Ti had virtually no suppression of T_c and H_{c2} despite the expectation of strong proximity coupling. This occurred because the proximity length of Ti was ~10 nm. The critical current density of a 45 nm / 15 nm multilayer exhibited matching effects. The 10/10 multilayer achieved J_c values comparable to those of a Nb47Ti tape conductor, reaching 5 kA/mm^2 at 5 T, 4.2 K. The 10/10 multilayer also approached the limit of adiabatic stability, which resulted in some J_c data being limited by a quench. Methods to improve the stability were discussed. Also, the critical current density data was compared to a previous result [7], and several differences in the experiments were found.

REFERENCES

[1] P.J. Lee and D.C. Larbalestier, Acta Metall., vol. 35, pp. 2523-2536, 1987.
[2] L.D. Cooley, P.J. Lee, and D.C. Larbalestier, Phys. Rev. B,vol. 53, pp. 6638-6652, 1996.
[3] C. Meingast, P.J. Lee, and D.C. Larbalestier, J. Appl. Phys., vol. 66, pp. 5962-5970, 1989.
[4] K. Matsumoto, Y. Tanaka, K. Yamafuji, M. Iwakuma, and T. Matsushita, IEEE Trans. Appl. Supercond., vol. 5, pp. 1362-1365, 1993.
[5] R.W. Heussner, C.B. Nunes, L.D. Cooley, and D.C. Larbalestier, IEEE Trans. Appl. Supercond., vol. 7, pp. 1142-1145, 1997.
[6] E. Kadyrov, A. Gurevich, and D.C. Larbalestier, Appl. Phys. Lett., vol. 68, pp. 1567-1569, 1996.
[7] J.D. McCambridge et al., IEEE Trans Appl. Supercond., vol. 7, pp. 1134-1137, 1997.
[8] W.H. Warnes, K.J. Faase, and N.A. Norris, Adv. Cryo. Eng. (Materials), vol. 42B, pp. 1143-1150, 1996.
[9] The bridge width reported in ref. [6], 50 μm, was in error. The correct value is 150 μm.
[10] L.D. Cooley, C.D. Hawes, and D.C. Larbalestier, to be published.
[11] Y. Kuwasawa, T. Tosaka, A. Uchiyama, S. Matuda, and S. Nakano, Physica C, vol. 175, pp. 187-191, 1991.
[12] L.D. Cooley, P.D. Jablonski, P.J. Lee, and D.C. Larbalestier, Appl. Phys. Lett., vol. 58, pp. 2984-2986, 1991.
[13] C.D. Hawes, L.D. Cooley, and D.C. Larbalestier, paper MFA-01, this conference.
[14] G. Stejic, A. Gurevich, E. Kadyrov, D. Christen, R. Joynt, and D.C. Larbalestier, Phys. Rev. B, vol. 49, pp. 1274-1288, 1994.
[15] S. Takahashi and M. Tachiki, Phys. Rev. B, vol. 33, pp. 4620-4631, 1986.
[16] S.H. Brongersma, E. Verweij, N.J. Koeman, D.G. deGroot, R. Griessen, and B.I. Ivlev, Phys. Rev. Lett., vol. 71, pp. 2319-2322, 1993.
[17] M.N. Wilson, Superconducting Magnets, Oxford: Clarendon, 1986.
[18] A.V. Gurevich and R.G. Mints, Rev. Mod. Phys.,vol. 59, pp. 941-999, 1987.