Pinning and Vortex Lattice Structure in NbTi Alloy Multilayers

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***Abstract*—We made thin film multilayers of $Nb_{0.37}Ti_{0.63}$/Nb and $Nb_{0.37}Ti_{0.63}$/Ti (d_{NbTi} = 14-27 nm and d_N = 4-11 nm) to examine geometries and materials relevant to flux pinning in commercial NbTi conductors. Samples were characterized by transport measurements between 4.2 K and T_c, in magnetic fields nearly parallel to the layers, up to 6 T. For some multilayers, pinning forces had a large peak at intermediate fields whose onset occurred near ~ 0.2 H_{c2}. We suggest this peak effect is caused by a change in the vortex lattice structure, driven by the strong intrinsic pinning. We have measured the highest pinning force density (113 GN/m^3 at 4.2 K and 5 T) ever achieved in the NbTi system.**

With the development of superconducting artificial pinning center (APC) wires [1], [2], the behavior of the vortex lattice in a periodic pinning system has become of renewed importance. To investigate the influence of structure and material on the ultimate pinning forces achievable in APC wires, we made thin film multilayers (MLs) of NbTi/Nb and NbTi/Ti. The guidance that multilayers can provide for wire makers is constrained somewhat by the richness of layered samples' behavior—the benefits gained from their quasi-2D nature are difficult (but not impossible [1]) to mimic on a larger scale. There have been a number of previous works which have investigated the superconducting properties of similar MLs [3], [4]. There are only a few other studies which also examine the flux pinning properties of Nb-alloy MLs at temperatures and geometries appropriate for comparison with APC wires [5]-[8].

Our fabrication and measurement techniques were published in detail earlier [8]. Briefly, we sputter-deposited 11 period $Nb_{0.37}Ti_{0.63}$/Nb and $Nb_{0.37}Ti_{0.63}$/Ti MLs onto (100) Si wafers at ~ 255 °C. The NbTi layer (referred to as the S layer [9]) had thickness d_S from 14-27 nm. The Nb and Ti layers (N layer [9] or barrier layer) had thicknesses d_N between 4-11 nm. Buffer and cap layers of Nb or Ti (as appropriate) were 50 nm thick; these were deposited to protect the top and bottom of the MLs and inhibit surface superconductivity[4]. Films were patterned by photolithography and reactive ion etching into wires 3 μm wide and 60 μm long. Sample geometries were confirmed by SEM and TEM. We made transport measurements of the critical current density J_c and the upper critical field H_{c2} as functions of temperature and the angle θ between the sample surface and the magnetic field B (see Fig. 1 inset). A total of 6 NbTi/Nb and 2 NbTi/Ti multilayer samples (see Table 1), as well as single films of NbTi, Nb, and Ti, were characterized in this study. A voltage criterion of 0.3 μV was used to determine J_c. Our sample stage could be rotated *in situ* with a precision of ~ 0.25° and an absolute accuracy of 1.5°.

In Fig. 1 we plot $J_c(\theta)$ for three samples: a 200 nm thick film of NbTi and multilayers B and H (see Table 1 for layer thicknesses). These data were taken at 4.2 K and 5 T. All three show cusps at $\theta = 0°$, but the J_c of both MLs is surprisingly large. The maximum J_c for the NbTi/Ti sample (H) is at $\theta = 0°$ and corresponds to a pinning force density $F_p = J_c B$ = 113 GN/m^3. This is the *highest* pinning force ever measured in the NbTi system [7], [10]. For thin ($d < \lambda$) samples, some enhancement of J_c in a parallel field is expected due to surface pinning [10]. The 200 nm NbTi film is thinner than the MLs, yet its J_c is negligible. Thus, surface enhancement of J_c is not an important effect for our multilayer samples.

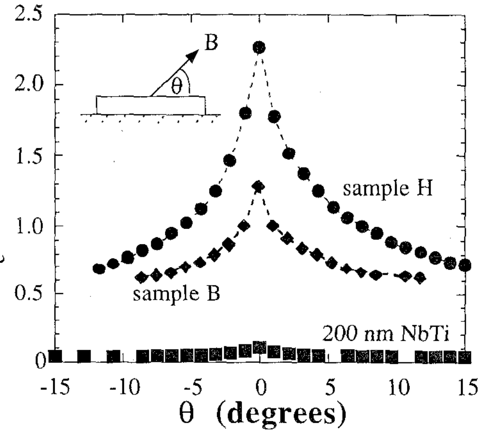

Fig. 1. The critical current density J_c versus the angle the applied magnetic field B makes with the sample surface θ for a 200 nm NbTi film and multilayers B and H.

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TABLE 1. NB-ALLOY MULTILAYER SAMPLE GEOMETRIES

Name	N layer	d_N (nm)	d_S (nm)	thickness[a] (nm)
A	Nb	4.0	20.1	365
B	Nb	5.7	20.6	390
C	Nb	7.7	20.8	415
D	Nb	10.4	20.1	435
E	Nb	9.0	14.2	355
F	Nb	8.8	26.9	495
G	Ti	9.0	20.6	425
H	Ti	5.2	19.4	370

[a] includes buffer and cap layer

The data in Fig. 2 were taken at 4.2 K and $\theta = 5.4°$ (to avoid sample heating at low fields, i.e. at large currents [5]). For these films, d_S is similar while d_N varies. In Fig. 2(a) we plot $J_c(B)$ for the NbTi film and samples D and G. The J_c of the NbTi and NbTi/Ti (G) samples are both decreasing with field. The NbTi/Nb sample (D) shows a strong non-monotonic field dependence of J_c in the range 1-4 T. In the inset of Fig. 2(a) are shown the corresponding $F_p(B)$. The pinning force density curve of sample D also exhibits non-standard behavior. This behavior persisted out to quite large angles $\theta \sim 60°$. This has been observed by others as well [5].

Typically, $F_p(B)$ for isotropic films is a dome-like function with a single maximum [11]. We see in the insets of Fig. 2 that the pinning force curves of the layered samples can have a shoulder at low fields or even two maxima. We define two fields which we will use to characterize the non-standard field dependence: the peak field B_{max} and the onset field B_{min}, which we have marked in Fig. 2(a). B_{max} is the field at maximum F_p. B_{min} is the inflection point or local minimum on the pinning force density curve, which corresponds closely with the local minimum of J_c. We choose F_p to characterize this peak effect, rather than J_c, because it is a more fundamental quantity and samples which have no minimum in J_c *do* have recognizable structure in F_p. Other groups have chosen to characterize the field dependence in superconducting MLs in terms of J_c [5], [6], [12]. The characteristic fields obtained by that method scale with those obtained from F_p, in general.

In Fig. 2(b) and its inset, we plot $J_c(B)$ and $F_p(B)$ for multilayers with thinner N layers: samples B and H. Now both samples show a pinning peak at intermediate fields. Comparing Fig. 2(a) and 2(b), as the Nb layer becomes thicker, $F_p(B_{max})$ increases. For Ti layers, the opposite occurs. For both N materials, B_{max} shifts to higher fields as d_N decreases. We present the structural dependences of the characteristic fields at 4.2 K and $\theta = 5.4°$ for our NbTi/Nb MLs in Fig. 3. In Fig. 3(a) we plot the dependence of the peak field B_{max} versus the N layer thickness d_N. We find that B_{max} scales weakly with $1/d_N$ and is independent of d_S, as indicated by the sample names on the figure. This contradicts a simple model of matching or commensurability between the vortex lattice and the pinning lattice to explain the peak in F_p [12], [13], [14]. In the case of geometric matching, the position of the peak would scale with $1/\Lambda^2$, where $\Lambda = d_S + d_N$ is the period of the S-N bilayer. Other workers have also observed a dependence of the peak position on $1/d_N$ [5]. Fig. 3(b) shows the dependence of the onset field B_{min} on the period Λ. B_{min} apparently scales with $1/\Lambda$, but we have investigated only a fairly narrow range of ML periods. More data need to be obtained to differentiate between $1/\Lambda$ and $1/\Lambda^2$ scaling. No other group has presented systematic data on B_{min}.

We present the temperature dependence of the upper critical field, as well as B_{max} and B_{min}, for a typical ML, sample C, in Fig. 4. Again, these data were taken at $\theta = 5.4°$. $H_{c2}(T)$ shows the 3D-to-quasi-2D dimensional crossover of the superconducting order parameter Δ occurring at T ~ 8.1 K [9]. This crossover is well-known in layered superconductors [3]-[6], [15]. In the quasi-2D regime below 8 K in Fig. 4, $H_{c2}(T) \approx H_{c2D} \sim (T—T_{3D\text{-}2D})^{1/2}$, where H_{c2D} is the upper critical field of an isolated thin ($d < \xi$) film and $T_{3D\text{-}2D}$ is the temperature below which the superconducting coherence length ξ normal to the layers becomes comparable to Λ [5], [9], [15]. Both characteristic fields decrease as the temperature is increased,

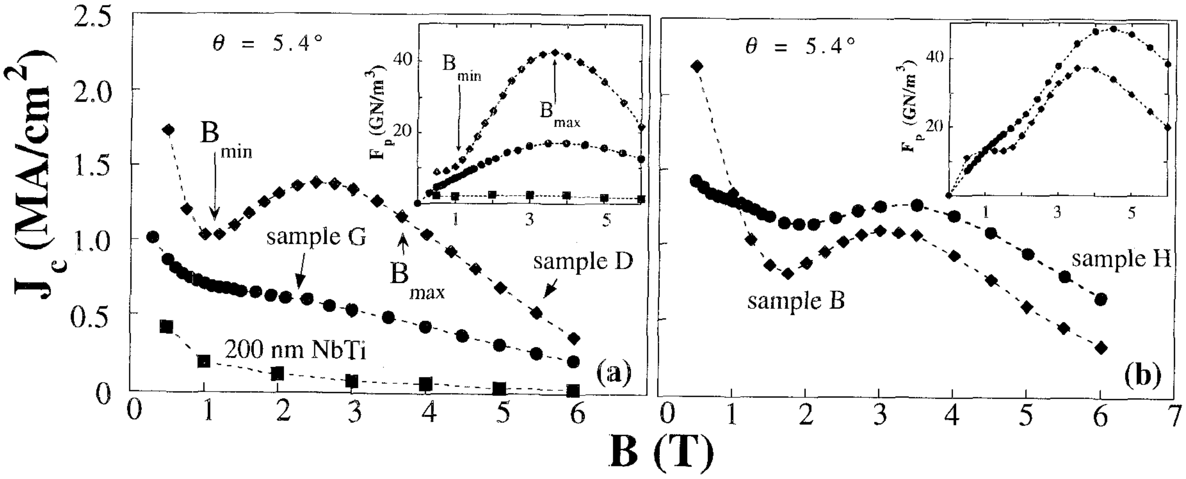

Fig. 2. Critical current density J_c versus applied field B at 4.2 K and $\theta = 5.4°$: (a) comparison of layered (Nb and Ti) and unlayered samples, (b) the influence of thinner Nb and Ti layers. Insets: the corresponding global pinning force densities F_p. The characteristic fields B_{max} and B_{min} are marked in (a).

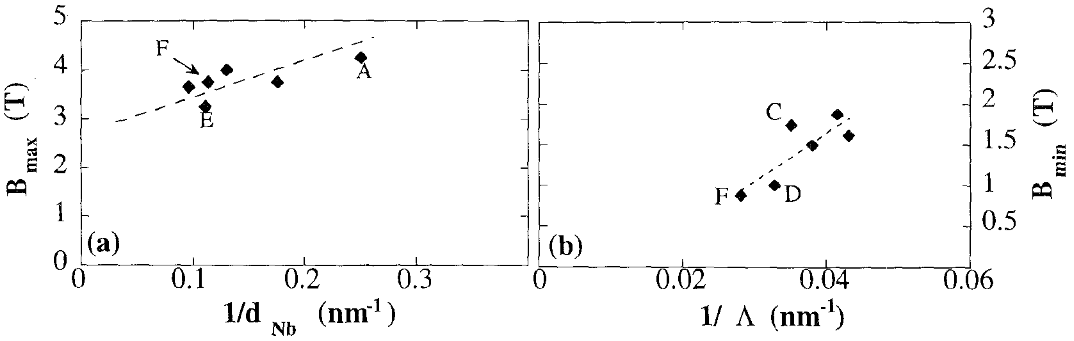

Fig. 3. The structural dependences of the characteristic fields at 4.2 K and $\theta = 5.4°$ for all the NbTi/Nb multilayers measured: (a) the peak field scales as $1/d_N$ and has no systematic dependence on d_S or period Λ, as indicated by the marked samples; (b) the onset field scales approximately as $1/\Lambda$. The lines are guides to the eye only.

but we can find no simple temperature dependence for the onset field B_{min}. We find that the peak field $B_{max} \approx 0.5H_{c2D} \sim (T—T_{3D\text{-}2D})^{1/2}$ for all the NbTi/Nb samples across the full temperature range we explored. Similar scaling has been seen by other workers [5]. The temperature dependence of both characteristic fields also contradicts a simple commensurability argument for the peak effect.

Tachiki and Takahashi proposed a model, the step-wise or kinked vortex lattice (SVL), to describe the vortex structure in layered superconductors [16]. Recent experimental work on Nb-alloy MLs suggests that the non-monotonic dependence of the pinning forces on applied magnetic field is related to the crossover between a straight (anisotropic Abrikosov) and an SVL configuration [5], [6], [8]. The appearance of a peak in the pinning force can be traced to the competition between the vortex-vortex and vortex-pin interactions [11]. The step-wise vortex lattice model does not take intervortex repulsion into account. In a typical system with strong, random pinning, this neglect of repulsion is justified: the shear and tilt energies of the flux lattice are much smaller than the pinning energy U_{pin}; usually the energy of uniaxial compression U_{comp} is much larger than U_{pin} and the vortices are also assumed to be incompressible.

When the magnetic field is aligned with the plane of the layers, the pinning energy can be extremely large. The variation of the order parameter Δ across the layers gives rise to a periodic intrinsic pinning potential [13], [16]. We claim the intrinsic pinning dominates the behavior of our samples—simple estimates of the elementary pinning force per unit length f_L of the interfaces between NbTi and Nb [17] are of the same order of magnitude as our measured values $f_L \approx F_p(B_{max})\xi_{NbTi}/n_L \approx 0.5$ µN/m, where $n_L = 1/\Lambda$, the number density of interfaces. The quasi-2D scaling of B_{max} seen in Fig. 4 also supports this argument. Pinning due to the intrinsic potential should scale with the variation of Δ across the layers: at $T_{3D\text{-}2D}$, Δ has weak variation across the sample and $U_{pin} \rightarrow 0$. The total pinning force also depends on the length of vortex which interacts with the pinner—in the field-aligned case this is essentially the sample size. Hence we find much higher pinning forces than can be obtained in the bulk, where the effective length of the pinners is a small fraction of the wire size [18]. This leads us to propose below an explanation for the mechanism underlying the non-standard dependence of the pinning forces on B in near-parallel fields that we and others have observed.

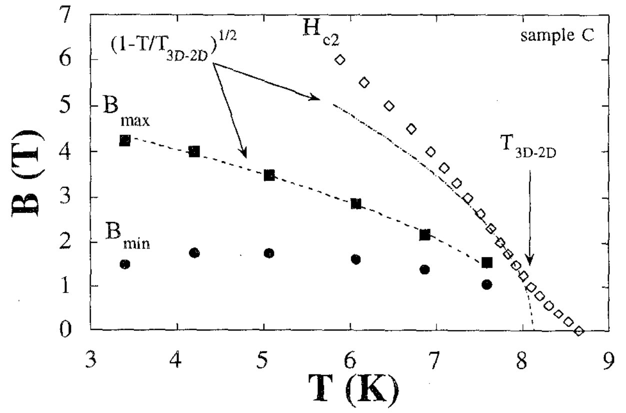

Fig. 4. The phase diagram of multilayer C at $\theta = 5.4°$. The upper critical field H_{c2}, the peak field B_{max}, and the onset field B_{min} are plotted as functions of temperature T. The dashed lines are fits to quasi-2D behavior $H_{c2D} \sim (T-T_{3D\text{-}2D})^{1/2}$, see text.

Our model to explain this peak effect begins with the dispersive nature of the compression modulus c_{11}: short wavelength compressions of the vortex lattice cost less energy than long wavelength ones. At low fields, the vortices in the multilayer are far apart and the compressive distortions necessary to form the SVL require high energy. As the field increases, U_{comp} drops at the same time that the pinning energy is increasing to its maximum. If U_{pin} becomes greater than U_{comp}, we expect a transition from a rigid to a soft, compressible vortex lattice—this is the postulated crossover between an anisotropic Abrikosov and step-wise vortex lattice which causes the pinning force to rise suddenly [5]. We identify the field at which this transition occurs between a rigid and a step-wise vortex lattice as the onset field B_{min}.

So far, we have been primarily discussing the origins of the peak effect in the pinning forces of the NbTi/Nb samples. For the NbTi/Ti MLs, the situation is slightly different: for sample H, with thin Ti layers ($d_N = 5.2$ nm), we observe a strong peak similar to (and exceeding) that seen in the NbTi/Nb samples (compare Fig. 2(a) and 2(b)). For sample G, with thicker Ti layers ($d_N = 9.0$ nm), no peak can be discerned and the overall pinning force density is lower (see Fig. 2(a)). We postulate that the sample with thicker Ti layers is too weakly coupled to sustain a vortex in the barrier [19], which would make the intrinsic pinning ineffective. Thus, a peak effect should only arise in a layered superconductor in which the S layers are strongly coupled through the N layers, either because both are superconductors or one is a normal metal not much thicker than the exponential decay length of the order parameter.

We fabricated $Nb_{0.37}Ti_{0.63}/Nb$ and $Nb_{0.37}Ti_{0.63}/Ti$ multilayers with geometries analogous to those of APC wires to better understand the roles of second-phase materials and structure in flux pinning. These materials and geometries have not been extensively studied before. We observed the expected 3D-to-2D crossover of the order parameter Δ in the upper critical field. In our measurements of critical currents in magnetic fields near parallel to the layers, we observed strong peak effects in the pinning forces as a function of field in the range 0.2-0.5 H_{c2}. This peak is caused by a change in the vortex lattice structure driven by the competition between the intrinsic pinning of the layers and the elastic forces between vortices. We have recorded the highest ever pinning force in the NbTi system, F_p = 113 GN/m^3 at 5 T and 4.2 K, in a NbTi/Ti multilayer.

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REFERENCES

[1] K. Matsumoto *et al.*, "Enhanced J_c properties in superconducting NbTi composites by introducing Nb artificial pins with a layered structure," *Appl. Phys. Lett.*, vol. 64, pp. 115-7, 1994.

[2] L. R. Motowidlo, B. A. Zeitlin, M. S. Walker, and P. Haldar, "Multifilament NbTi with artificial pinning centers: the effect of alloy and pin material on the superconducting properties," *Appl. Phys. Lett.*, vol. 61, pp. 991-3, 1992.

[3] for example Y. Kuwasawa *et al.*, "Observation of anomalous transition in the upper critical fields of $Nb/Nb_{0.5}Zr_{0.5}$ multilayers," *Physica C*, vol. 165, pp. 173-8, 1990; M. G. Karkut *et al.*, "Anomalous upper critical fields of superconducting multilayers: verification of the Takahashi-Tachiki effect," *Phys. Rev. Lett.*, vol. 60, pp. 1751-4, 1988; Y. Obi, M. Ikebe, Y. Muto, and H. Fujimori, "Upper critical fields of NbTi based multilayered materials," *Jap. J. Appl. Phys.*, vol. 26-3, pp. 1445-6, 1987.

[4] W. Maj and J. Aarts, "Outer layers determine the parallel critical field of a superconducting multilayer," *Phys. Rev. B*, vol. 44, pp. 7745-8, 1991.

[5] P. Koorevaar, W. Maj, P. H. Kes, and J. Aarts, "Vortex-lattice transition in superconducting NbZr/Nb multilayers," *Phys. Rev. B*, vol. 47, pp. 934-43, 1993.

[6] T. Nojima, M. Kinoshita, S. Nakano, and Y. Kuwasawa, "Transport critical current density and dimensional crossover in superconducting NbZr/Nb multilayers," *Physica C*, vol. 206, pp. 387-92, 1993.

[7] E. Kadyrov, A. Gurevich, and D. C. Larbalestier, "Very high critical current densities in Nb47%Ti multilayers with a planar copper flux pinning nanostructure," *Appl. Phys. Lett.*, vol. 68, pp. 1567-9, 1996.

[8] J. D. McCambridge, "The superconducting properties of Niobium-Titanium alloy multilayers," Yale University thesis, unpublished, 1995; J. D. McCambridge *et al.*, "Flux pinning in NbTi/Nb multilayers," *IEEE Trans. Appl. Supercond.*, vol. 5, pp. 1697-1700, 1995

[9] S. Takahashi and M. Tachiki, "Theory of the upper critical field of superconducting superlattices," *Phys. Rev. B*, vol. 33, pp. 4620-31, 1986; "New phase diagram in superconducting superlattices," *ibid.*, vol. 34, pp. 3162-4, 1986.

[10] G. Stejic *et al.*, "Effect of geometry on the critical currents of thin films," *Phys. Rev. B*, vol. 49, pp. 1274-88, 1994.

[11] A. I. Larkin and Yu. N. Ovchinnikov, "Flux pinning in type II superconductors," *J. Low Temp. Phys.*, vol. 34, pp. 409-28, 1979.

[12] H. Raffy and E. Guyon, "Dependence of critical current and field of periodically modulated superconducting alloys on modulation amplitude," *Physica*, vol. 108B, pp. 947-8, 1981; H. Raffy, J. C. Renard, and E. Guyon, "Critical currents and pinning effect in superconducting alloy films spatially modulated in concentration," *Sol. State Comm.*, vol. 11, pp. 1679-82, 1972.

[13] S. H. Brongersma *et al.*, "Series of maxima in the field dependent magnetic moment of layered superconductors," *Phys. Rev. Lett.*, vol. 71, pp. 2319-22, 1993.

[14] S. Ami and K. Maki, *Prog. Theor. Phys.*, vol. 53, p. 1, 1975.

[15] S. T. Ruggiero, T. W. Barbee, and M. R. Beasley, "Superconducting properties of Nb/Ge metal semiconductor multilayers," *Phys. Rev. B*, vol. 26, pp. 4894-4908, 1982; R. A. Klemm, A. Luther, and M. R. Beasley, "Theory of the upper critical field in layered superconductors," *Phys. Rev. B*, vol. 12, pp. 877-91, 1975.

[16] M. Tachiki and S. Takahashi, "Anisotropy of critical current in layered oxide superconductors," *Sol. State Comm.*, vol. 72, pp. 1083-6, 1989; "Strong vortex pinning mechanism in high-T_c oxide superconductors," *ibid.*, vol. 70, pp. 291-5, 1989.

[17] G. Stejic, "The mean-field flux pinning theory," University of Wisconsin-Madison thesis, unpublished, 1993; G. Stejic *et al.*, "Numerical calculation of flux pinning by α-Ti precipitates in Nb-Ti," *Supercond. Sci. Tech.*, vol. 5, p. S97, 1992.

[18] C. Meingast, P. J. Lee, and D. C. Larbalestier, "Quantitative description of a high J_c NbTi superconductor during its final optimization strain: I. Microstructure, T_c, H_{c2}, and resistivity," *J. Appl. Phys.*, vol. 66, pp. 5962-70, 1989

[19] A. Gurevich and L. D. Cooley, "Anisotropic flux pinning in a network of planar defects," *Phys. Rev. B*, vol. 50, pp. 13563-76, 1994.