SCALING LAWS FOR THE CRITICAL CURRENT DENSITY
OF NbN FILMS IN HIGH MAGNETIC FIELDS*

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1992 Applied Superconductivity Conference, Chicago, IL, August 24-28, 1992

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*Work supported by the University Funding Council U.K., Science and Engineering Council, U.S. Department of Energy BES-MS under contract #W-31-109-ENG-38, and Office of Fusion U.S.A.

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ABSTRACT

We have measured the critical current density (J_c) of two NbN films (500 Å and 1550 Å thick) as a function of temperature in magnetic fields up to 25 Tesla using transport measurements. In both films, the functional form of the volume pinning force F_p obeys the Fietz - Webb scaling law throughout the entire magnetic field and temperature range such that:

$$F_p = J_c \times B = \alpha B_{c2}^m(T) b^{1/2} (1-b)^2 = \alpha' (1-T/T_c)^m b^{1/2} (1-b)^2$$

where α and α' are constants dependent on the film, $B_{c2}(T)$ is the upper critical field, $b = B/B_{c2}(T)$ is the reduced magnetic field, T_c is the critical temperature and we find $m = 2.7 \pm 0.1$. Over a limited range of magnetic fields close to $B_{c2}(T)$, we can approximate this functional form by:

$$F_p = \beta B_{c2}^M(T) b (1-b)^2 = \beta' (1-T/T_c)^M b (1-b)^2$$

where β and β' are constants and we find $M = 2.6 \pm 0.2$. Values of J_c derived from D.C. magnetisation data obtained using Bean's model show qualitative agreement with the transport measurements throughout the superconducting phase. Despite the marked granularity in the microstructure of these films, we interpret our results as evidence that a flux pinning mechanism determines the transport current density in NbN films in high magnetic fields.

INTRODUCTION

In many low temperature superconductors which have been optimised for high J_c in high magnetic fields, a single microstructural or compositional feature such as the grain boundary density or the second phase material present has been identified as the dominant factor that determines J_c (1-3). Although a detailed understanding of the pinning mechanism in these materials is still not complete, general pinning models have been developed using the simple correlations between the electromagnetic properties and the physical properties. These models have facilitated optimising J_c for high field applications.

In polycrystalline ceramic oxide superconductors, no equivalent description is available. It is clear that the electromagnetic properties, and particularly the high field transport properties are sensitive to changes in fabrication that are not well understood (4). Although descriptions of the flux motion at J_c in high magnetic fields and the type of mechanism that determines J_c have been proposed (5-7), these descriptions have yet to incorporate the relationship between the critical current density and the physical properties of the

superconductor. Such a description would aid both the development of models that would explain the origin of the mechanism that determines J_c in these materials and the optimisation of J_c for high field applications.

In this work we present J_c data for two niobium nitride films. This material shows the isotropy and relatively long coherence length typical of low temperature superconductors while also having both strongly granular physical properties and large inter- and intragranular J_c typical of the oxides (8,9). Hence it serves as a useful model system, intermediate between the low temperature superconductors and the ceramic oxide superconductors which may aid in our development of pinning models for the oxides. In addition, there is the well established technological interest in this material for its potential use in fusion applications because of the insensitivity of J_c in NbN to strain (10) and radiation damage (11,12).

Measurements of J_c throughout the superconducting phase from 2K up to the critical temperature (T_c) in magnetic fields up to 25 Tesla are presented. These data provide information as to the mechanism limiting J_c in NbN thin films.

Fig 1: The critical current density measured at an electric field criterion of 1 microvolt per cm as a function of magnetic field and temperature for the 500 Å film.

SAMPLE PREPARATION

The NbN thin films were produced using standard D.C. reactive magnetron sputtering techniques (13). Nb was reactively sputtered in a Argon/Nitrogen atmosphere at a rate of $\sim 2\text{nm.s}^{-1}$ onto a sapphire substrate maintained at $\sim 300\text{C}$. Two films were produced, one 500 Å thick the other 1550 Å. The films were patterned using standard photolithographic techniques to produce tracks of length 6.5mm and width 1mm. The films were prepared for electrical measurements by ultrasonically soldering indium pads to the samples.

A detailed investigation of similar films using T.E.M. has been completed (14). The analysis suggests that the films consist of predominantly columnar FCC grains of about 10-20nm. The grains themselves are highly defective of nitrogen. The grain boundaries, unlike metals, are about 1.5nm thick - probably of insulating material (15).

APPARATUS

Standard four-terminal voltage-current (V-I) data were generated as a function of magnetic field and temperature. The data at 4.2K, 3K and 2K were obtained by directly immersing the sample in liquid helium. Above 4.2K, a variable temperature insert was used. In zero field the temperature was fixed using a calibrated carbon glass thermometer. A magnetic-field-independent strontium titanate thermometer was used in closed loop with a heater to keep the temperature constant when the magnetic field was applied and the V-I characteristic measured. The hybrid magnet at the Francis Bitter National High Field Laboratory was used to provide the magnetic fields up to 25T.

EXPERIMENTAL RESULTS

In figs 1 and 2, the critical current density measured as a function of magnetic field and temperature at an electric field criterion of one microvolt per cm is presented for the two NbN films. The electric field criterion was chosen to be as

Fig 2: The critical current density measured at an electric field criterion of 1 microvolt per cm. as a function of magnetic field and temperature for the 1550 Å film.

low as possible while retaining good signal-to-noise. The magnitude of J_c in the two films is similar and characteristic of high quality material. It can be seen that by using the hybrid magnet we have been able to make measurements of J_c up to $B_{c2}(T)$ throughout the entire temperature range from the critical temperature down to 2K for both films.

The shape of the V-I characteristic is often described by an empirical equation of the form:

$$E = \beta J^n$$

where E is the electric field, J is the current density and β and n are constants (16). The index n is used as a simple diagnostic to determine the homogeneity of the sample and in particular whether the material has been damaged. In figs 3 and 4 the value of the index n defined using equation 1 over the electric field range 1 to 10 microvolts per cm is presented for both films. The functional form and magnitude of n is typical of high J materials for technological use, having a value of about 20-30 at 10T., 4.2K and decreasing as either the temperature or the magnetic field increases.

An alternative description of the V-I characteristic is provided by considering the differential resistivity (ρ) above J_c . In figs 5 and 6, the differential resistivity (ρ_d) for each of the films is presented as a function of field and temperature, where:

$$\frac{\partial E}{\partial J} = \rho_d$$

ρ_d is calculated over the electric field range from 5 microvolts per cm. It should be noted that at every temperature, the data are consistent with ρ_d being proportional to B at low magnetic fields.

We decided to investigate whether the current path is percolative by measuring how the critical current density depends on the relative orientation of the macroscopic current direction and the applied field. In fig 7, J_c data

Fig 3: The index (n) calculated over the electric field range 1-10 microvolts per cm as a function of magnetic field and temperature for the 500 Å film.

Fig 4: The index (n) calculated over the electric field range 1 - 10 microvolts per cm. as a function of temperature for the 1550 Å thick film.

are presented for three configurations of J and B . These data are all taken at 4.2K on the 500 Å thin film. It can be seen that the field at which the current drops to zero (ie $B_{c2}(T)$) does not depend on the orientation of the applied field. When J is orthogonal to B , J_c is a function of field alone independently of whether the field is orthogonal or parallel to the surface of the film. Finally when there is no net Lorentz force operating (ie $J \parallel B$), J_c is higher than when a macroscopic Lorentz force operates by typically 50%. A similar enhancement of J_c to that found at 4.2K was also measured at 3K in the 500 Å film.

In addition to the transport measurements, a series of D.C. magnetisation measurements on the 500 Å thin film were completed. A sample of area 1mm x 1mm was cut from the track in the film and the magnetic moment measured as a function of field and temperature. A critical current density was calculated using Bean's model (17) assuming that the currents induced flowed over the whole dimensions of the sample. In fig 8, a comparison between this magnetically determined critical current density and the transport measurements is presented. It can be seen that there is qualitative agreement between J_c found in the two types of measurement.

Fig 5: The differential resistivity calculated over the electric field range 1 - 5 microvolts per cm as a function of magnetic field and temperature for the 500 Å film.

Fig 6: The differential resistivity calculated over the electric field range 1 - 5 microvolts per cm. as a function of magnetic field and temperature for the 1550 Å film.

We intended to measure how the magnetisation scaled with sample dimensions but were not successful. The NbN was cut using a diamond saw. The magnetic moment dropped precipitously as the sample dimensions dropped below 1mm² which we attributed to damaging the sample during the cutting procedure.

ANALYSIS OF RESULTS

When the volume pinning force F_p obeys the Tietz Webb scaling law (18), where:

$$F_p = J_c \times B = \alpha \cdot B_{c2}^\gamma(T) \cdot f(b) \quad - \text{eqn.}$$

α and γ are constants, $B_{c2}(T)$ is the upper critical field and $f(b)$ is a function of the reduced magnetic field ($= B/B_{c2}(T)$), this provides evidence that the critical current density is limited by a flux pinning mechanism. We shall consider whether either a shearing (pinning) mechanism or a core pinning mechanism can describe our data.

If a shearing mechanism limits J_c then $f(b) = b^{1/2} \cdot (b)^2$. In this case we can write the expression for J_c in the

Fig 7: The orientation dependence of the critical current density as a function of field at 4.2K in high magnetic fields for the 500 Å film.

Fig 8: A comparison between the critical current density derived from magnetic measurements with that measured using transport measurements on the 500 Å thick film.

form:

$$F_p = J_c \times B = \alpha \cdot B_{c2}^m(T) b^{1/2} \cdot (1-b)^2 \quad \text{eqn. 2}$$

where m is a constant. This can be rewritten:

$$J_c^{1/2} B^{1/4} = G \cdot (B_{c2}(T) - B).$$

where $G = \alpha^{1/2} \cdot B_{c2}^{(m-3/2)/2}(T)$. It is clear that if J_c obeys the Kramer dependence then $J_c^{1/2} B^{1/4}$ is a linear function of the magnetic field. In figs 9 and 10, the data in figs 1 and 2 have been replotted as $J_c^{1/2} B^{1/4}$ versus B (a Kramer plot). To within the accuracy of our measurements, the data taken at each temperature are linear for both films and hence obey the Kramer functional form.

Alternatively if a core pinning mechanism limits J_c , $f(b)$ is of the form: $b \cdot (1-b)^2$. In this case eqn. 1 can then be written:

$$F_p = J_c \times B = \beta \cdot B_{c2}^M(T) \cdot b \cdot (1-b)^2 \quad \text{eqn. 3}$$

where β and M are constants. It should be noted that eqn. 2 and eqn. 3 have the same magnetic field dependence

Fig 9: A Kramer plot for the 500 Å film

Fig 10: A Kramer plot for the 1550 Å film.

close to $B_{c2}(T)$, since in both equations in the high reduced magnetic field limit $f(b)$ is proportional to $(1-b)^2$. Equation 3 can be rewritten:

$$J_c^{1/2} = \beta^{1/2} \cdot B_{c2}^{(M-3)/2}(T) \cdot (B_{c2}(T) - B)$$

In figs 11 and 12, the data in figs 1 and 2 have been replotted as $J_c^{1/2}$ versus the applied field. It can be seen although the data in these plots are reasonably linear at fields close to $B_{c2}(T)$, at low reduced fields the data are not linear in magnetic field. Hence our data are best described within the flux shearing framework where F_p is proportional to $b^{1/2}$ at low fields rather than the core pinning model where F_p is proportional to b .

By extrapolating $J_c^{1/2} B^{1/4}$ linearly to zero we can determine a value for $B_{c2}(T)$ at each temperature. In fig 1 the values of $B_{c2}(T)$ derived for both films are presented as a function of temperature. It should be noted that $B_{c2}(T)$ is a linear function of temperature throughout the entire range of our measurements in magnetic fields up to 25 Tesla. Hence our NbN films show no paramagnetic limiting at reduced temperatures as low as 15% of T_c (20).

Values of G have been calculated from the gradient of the straight lines in the Kramer plots (figs 9 and 10). The uncertainty in the values of G calculated increases from

Fig 11. $J_c^{1/2}$ as a function of field and temperature for the 500 Å film.

Fig 12. $J_c^{1/2}$ as a function of field and temperature for the 1550 Å film.

about 3% at low temperatures to about 6% close to T_c .

Rewriting the expression for G , we have:

$$\log_{10}(G) = 0.5 \log_{10}(\alpha) + 0.5(m-2.5)\log_{10}(B_{c2}(T))$$

Fig 14 is a plot of $\log_{10}(G)$ versus $\log_{10}(B_{c2}(T))$ for both films. The gradient of the two (unlabelled) solid lines through the data, which serve as guides to the eye, depend only on the exponent m . From these data we find $m = 2.7 \pm 0.1$ for both films. Also plotted on the graph are solid lines showing what to expect if $m = 2.5$ or $m = 3$. It should be noted that the value of α (determined from the extrapolated value of $\log_{10}(G)$ when $\log_{10}(B_{c2}(T))$ is zero) is about 10% higher for the 500 Å film than the 1550 Å film. Hence, we have found for both films that the volume pinning force throughout the entire field range is of the form:

$$F_p = J_c \times B = \alpha \cdot B_{c2}^{2.7}(T) b^{1/2} \cdot (1-b)^2 \quad \text{eqn. 4}$$

which can be written in the form:

$$F_p = \alpha^* (1-T/T_c)^{m/2} b^{1/2} \cdot (1-b)^2$$

where α and α^* are constants.

Although all our data can be described by eqn. 4, we

Fig 13: The upper critical field as a function of temperature for the two NbN films.

Fig 14: The prefactor G as a function of upper critical field for the two NbN films.

have also completed a similar analysis to that outlined above using the data generated on the 1550 Å film with the functional form given by equation 3. For this film we have found:

$$F_p = \beta \cdot B_{c2}^M(T) b \cdot (1-b)^2 \\ = \beta^* (1-T/T_c)^M b \cdot (1-b)^2 \quad \text{Eqn 5.}$$

where β and β^* are constants and we find $M = 2.6 \pm 0.1$. Equation 5 can also be used to describe the data for the 500 Å film close to $B_{c2}(T)$.

DISCUSSION

Flux pinning in NbN.

The marked granularity in the microstructure of these NbN films opens the possibility that the grains may be decoupled electromagnetically and hence pair-breaking may occur in the grain boundary region. Nevertheless, despite

the thick grain boundaries of non-superconducting composition, we have found that the functional form of J_c obeys the Fietz-Webb scaling law. This provides strong evidence that J_c is limited by flux pinning and any decoupling which occurs at the grain boundaries is not sufficient to limit J_c .

In thin films, an obvious candidate for the source of the pinning is the surface. However the orientation measurements shown in fig 7 demonstrate that when J is orthogonal to B , J_c does not depend on whether the applied field is parallel or orthogonal to the surface of the film. Hence we expect that bulk pinning is primarily responsible for high J_c rather than surface pinning.

The differential resistivity data (figs 5 and 6) are consistent with being proportional to the magnetic field at fixed temperature in the low field limit. This is a similar dependence to both the flux flow resistivity (21) and ρ_i found experimentally in other high J_c low temperature superconductors and hence also suggests that bulk pinning is operating (16,22,23). This dependence can be contrasted with a granular material where the differential resistivity is independent of magnetic field at fixed temperature (24).

From microstructural analysis of similar films to these, two potential sources of bulk pinning in these films have been identified: grain boundary pinning and intragranular pinning by nitrogen defects (12,14). The rather small increase in J_c when the macroscopic current direction is parallel to the applied field (fig 7) suggests that locally the current path is highly percolative. This weak dependence on the orientation of the field with respect to the macroscopic current direction is characteristic of a large variation in the local pinning force throughout the sample. Hence our data is consistent with both an inter- and intragranular contribution to the pinning.

Although there is quantitative agreement between the magnetic measurements of J_c and the transport measurements (fig 8), there is a large difference between the grain size ($\sim (100\text{nm})^2$) and the sample size ($\sim (1\text{mm})^2$) in our measurements. This difference implies that the intragranular current would have to be three orders of magnitude higher than the intergranular current density to increase the observed magnetic moment by only 10%. Hence, even if the intragranular J_c were close to the depairing limit, we would probably not detect it without additional measurements characterising how the magnetisation scales with size of the sample. As such, we leave as an open question the relative sizes and importance of the inter- and intragranular pinning.

Close to $B_{c2}(T)$, our data can be formally described using $f(b) = b \cdot (1-b)^2$. This facilitates a quantitative comparison between our data and other materials where core pinning operates. However the most complete description of our data throughout the entire magnetic field and temperature range for both films is provided using the Kramer dependence. Within this framework, the value of the exponent m is significantly larger than that found in materials where the grain boundaries are the sole source of pinning (eg $(\text{NbTa})_3\text{Sn}$ where $m = 2$ (25)) and hence provides evidence that the pinning mechanism is different. In conclusion, we need a more detailed under-

standing of the inter- and intragranular pinning in NbN before we can explain the functional form we have found.

NbN for high field applications

The high values for J_c and the index n we have found in high magnetic fields are characteristic of high quality NbN thin films. The higher value of α found in the 500 Å film is attributed to the higher grain boundary density. The higher critical parameters T_c and $B_{c2}(T)$ in the 1550 Å film are attributed to the longer annealing in nitrogen during fabrication and hence lower defect density within the grains. We expect that J_c would be increased yet higher in these films if one could reduce the grain size and/or decrease the nitrogen defect density. However too date, marked improvements in the properties of the single layer NbN films presented in this work have not been achieved. Furthermore, given the lack of evidence for any granularity, we do not expect to increase the J_c of these films by improving the structural order or off-stoichiometry at the grain boundaries. In practice it has been found that a markedly different approach has proved the best way to achieve larger J_c in high fields namely artificial multilayers which include artificial pinning layers (26).

CONCLUDING COMMENTS.

We have found that the volume pinning force obeys the Fietz-Webb scaling law which is strong evidence that a flux pinning mechanism limits J_c . More specifically:

$$\begin{aligned} F_p &= J_c \times B = \alpha \cdot B_{c2}^m(T) b^{1/2} \cdot (1-b)^2 \\ &= \alpha^* (1-T/T_c)^m b^{1/2} \cdot (1-b)^2 \end{aligned}$$

where α and α^* are constants and $m = 2.7 \pm 0.1$.

Over a limited range of magnetic fields close to $B_{c2}(T)$, we can approximate this functional form by an equation of the form:

$$\begin{aligned} F_p &= \beta \cdot B_{c2}^M(T) b \cdot (1-b)^2 \\ &= \beta^* (1-T/T_c)^M b \cdot (1-b)^2 \end{aligned}$$

where β and β^* are constants and we find $M = 2.60 \pm 0.15$

Above J_c the current flow locally is highly percolative and flux moves throughout the entire sample. The detailed description of the pinning mechanism is not clear. We suggest that there is both an intra- and intergranular contribution to the pinning which we attribute to the nitrogen defects within the grains and the grain boundaries respectively.

ACKNOWLEDGEMENTS

These electromagnetic measurements were made by DPH while he was a post-doctoral research scientist at The Applied Superconductivity Center, Madison, Wisconsin. The authors wish to thank Prof. D.C. Larbalestier for his considerable contribution to this work. In addition the authors acknowledge the contribution of Dr. D.W. Capone II for making the samples used in this study and B. Brandt and L. Rubin at The Francis Bitter National Magnet Laboratory. This work has support from the University Funding Council U.K., the Science and Engineering Council and the Department of Energy and Office of Fusion U.S.A.

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