CONF-800587--4

BNL-27964

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INTRODUCTION

With the increasing importance of multifilamentary Nb_3Sn conductors for technological uses such as for the production of very high magnetic fields in fusion magnets, means of improving the superconducting critical current density J_c at very high magnetic fields (H > 10 tesla) have been sought intensively, and it has been found that metallurgical factors such as heat treatment conditions,[1] alloying additions,[2] and mechanical strains[3] can strongly influence the critical current density. The correlation of changes in J_c with such metallurgical variations in the Nb_3Sn wires has been facilitated by the use of scaling laws for magnetic flux pinning in hard superconductors, and the scaling law developed by Kramer[4] has been used frequently.[5] We have found in the course of our investigations of the properties of monofilamentary Nb_3Sn wires produced by the "bronze process" that the magnetic field dependence of J_c at high fields can qualitatively be characterized well by Kramer's scaling law. However, when a detailed comparison of the scaling law and available experimental results was made, we found serious inconsistencies in the values of the parameters which appear in the scaling equation. In this article, we will point out those instances where the equation appears to work well and other cases where the use of the equation leads to unrealistic results.

*Work performed under the auspices of the U.S. Department of Energy.

THE SCALING LAW

The magnetic flux-pinning strength, F_p, in the high magnetic field regime is given by Kramer as[4]

$$F_p = |\vec{J}_c(H) \times \vec{H}| = K_s h^{1/2}(1-h)^2 \quad (1)$$

where $h = H/H_{c2}$ and $K_s = 0.56 H_{c2}^{5/2} \kappa_1^{-2}(1-a_o\sqrt{\rho})^{-2}$, and κ_1 is the Ginsburg-Landau constant ($\kappa_1 = H_{c2}/\sqrt{2}H_c$), a_o is the flux lattice spacing ($a_o = (\phi_o/H)^{1/2}$) and ρ is the density of the flux-pinning sites. This equation was derived assuming that flux pinning could be described by two régimes: at low fields flux motion occurs primarily by unpinning, whereas at high fields flux motion occurs by synchronous shear of the flux line lattice around pins too strong to be broken. Furthermore, the equation describes the dynamic pinning force produced by a series of pinning planes, each of which consists of a series of line pins, and which lie parallel to the Lorentz force.

In the past, the scaling law has often been tested by plotting experimental data as (F_p/F_{pmax}) vs h or F_p vs $h^{1/2}(1-h)^2$. A difficulty associated with such plots is the determination of a consistent value of the upper critical field H_{c2} needed to calculate the reduced magnetic field h appearing in Eq. (1). Also, when both axes are normalized it is not easy to make a critical comparison.

For studying the high field behavior of J_c or F_p, it is found that another form of Eq. (1) is more convenient and useful.[6,7] Simple algebraic manipulation of Eq. (1) yields:

$$J_c^{1/2} H^{1/4}(1-a_o\sqrt{\rho}) = 0.7\kappa_1^{-1}(H_{c2}-H) \quad . \quad (2)$$

Now, since the right-hand side of Eq. (2) is linear in H, the equation can be used to determine H_{c2} by plotting the left-hand side against H and extrapolating. In most cases, at sufficiently high field $a_o\sqrt{\rho} << 1$, and $J_c^{1/2}H^{1/4}$ is linear in H over a reasonably wide range of H; and thus H_{c2} is obtained simply by linear extrapolation and the Ginzburg-Landau parameter κ_1 is obtained from the slope of the plot without adjustable parameters. In the following section, the applicability of the scaling law, in the form of Eq. (2), is examined for bronze processed Nb_3Sn wires with regard to: 1) the linear dependence of $J_c^{1/2}H^{1/4}$ on H, and 2) the values of κ_1 determined from the slope.

EXPERIMENTAL RESULTS AND DISCUSSION

In order to examine the applicability of the scaling law, various experimental results for critical current densities in the bronze-processed wires are used. The details of fabrication for the wires and tapes are discussed elsewhere.[1] All of the measurements for J_c were made at the National Magnet Laboratory using a 19 T and a 23 T Bitter coil and at 4.2 K. The criteria for J_c was $\sim 1\ \mu V/2$ cm in most cases.

First, J_c data for a composite monofilamentary wire with a matrix-to-core ratio of ~ 15 and with a heat treatment of 725°C for 120 h are plotted in Fig. 1 as suggested by Eq. (2), illustrating

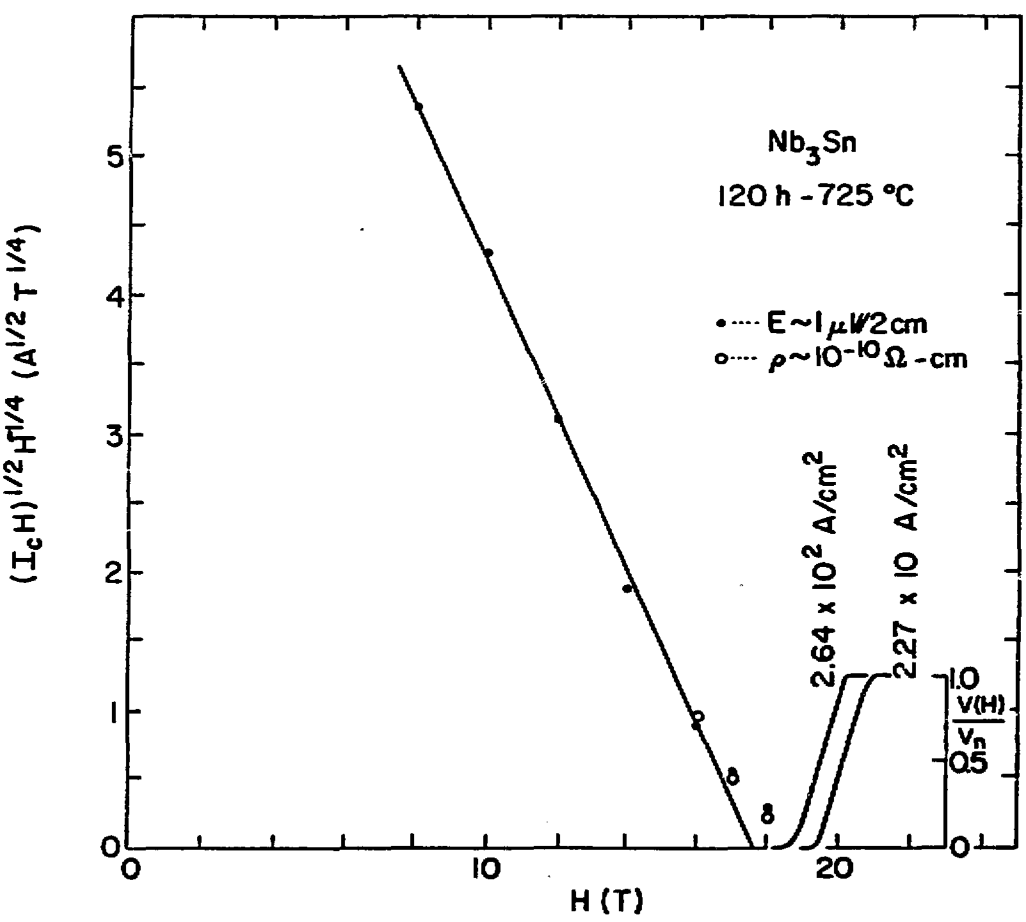

Fig. 1. A plot for $J_c^{1/2}H^{1/4}$ vs H for a monofilamentary Nb_3Sn wire which was heat treated at 725°C for 120 h indicating the extrapolated H_{c2} and the low current density H_{c2}.

its use in analyzing the magnetic field dependence of the critical current density. Here it was assumed that $a_o\sqrt{\rho} << 1$ in calculating the left-hand side of Eq. (2), and this assumption seems to be justified since the plot is a straight line with H over a wide range in magnetic field (∿8 to ∿16 T). This implies that the distance between the pinning sites in this wire is considerably larger than the flux lattice spacing, a_o. Therefore a measure of the upper critical field H_{c2} can be obtained by the linear extrapolation of the straight segment of the plot. This value of H_{c2} is not the magnetic field where superconductivity vanished totally from the wire, as may be seen in the figure. The experimental data deviate from a straight line near the critical field. In fact, superconductivity persists to considerably higher values (∿2 T) than the H_{c2} as determined by linear extrapolation. This is probably due to inhomogeneities in the Nb_3Sn such as a composition variation across the Nb_3Sn layer or a variation in the strains due to the matrix.[3] It is also possible that the bulk of the Nb_3Sn still carries superconducting currents beyond the extrapolated H_{c2}, and that the dependence of J_c on H does not follow Eq. (2) in that region, i.e., the scaling law fails here. Although the values of H_{c2} obtained by extrapolation will differ from those determined with other criteria, such as the midpoint of the transition from the normal to the superconducting state, H_{c2} as determined by this method will be consistent with Eq. (2) for the purpose of examination of the validity of the scaling law. Thus, in the following discussion, the values of H_{c2} quoted were all determined by the linear extrapolation method.

In the majority of cases, our measurements on monofilamentary Nb_3Sn wires show that J_c varies with magnetic field as described by Eq. (2) with $a_o\sqrt{\rho} << 1$, and thus yield linear plots of $J_c^{1/2}H^{1/4}$ versus H (except very near H_{c2}) as shown in Fig. 1. However in several instances, such simple quasi-linear plots were not obtained, and we believe such deviations fall into two categories.

In the first category, a plot of $J_c^{1/2}H^{1/4}$ vs H yields a plot with concave-up curvature. We believe that such curvature results from the erroneous assumption that $a_o\sqrt{\rho} << 1$. This behavior is illustrated in Figs. 2 and 3 which show data for a "bronze-processed" Nb_3Sn wire which was electron irradiated during a 500°C heat treatment[8] and for an "in-situ processed" wire, heat treated at 550°C for 6 days,[9] respectively. As shown in the figures, in both cases the data can be made to produce a wide region of linearity in the plot if the $(1-a_o\sqrt{\rho})$ term is included. Also in both cases, the selection of $(10^3\ \text{Å})^{-1}$ for the value of $\sqrt{\rho}$ resulted in a linear dependence on H. This value was found to give a better straight line fit than is obtained with $(500\ \text{Å})^{-1}$ or $(2000\ \text{Å})^{-1}$. It is interesting to note that in these wires the grain size of Nb_3Sn is ∿400Å[9] and yet the best fit for the

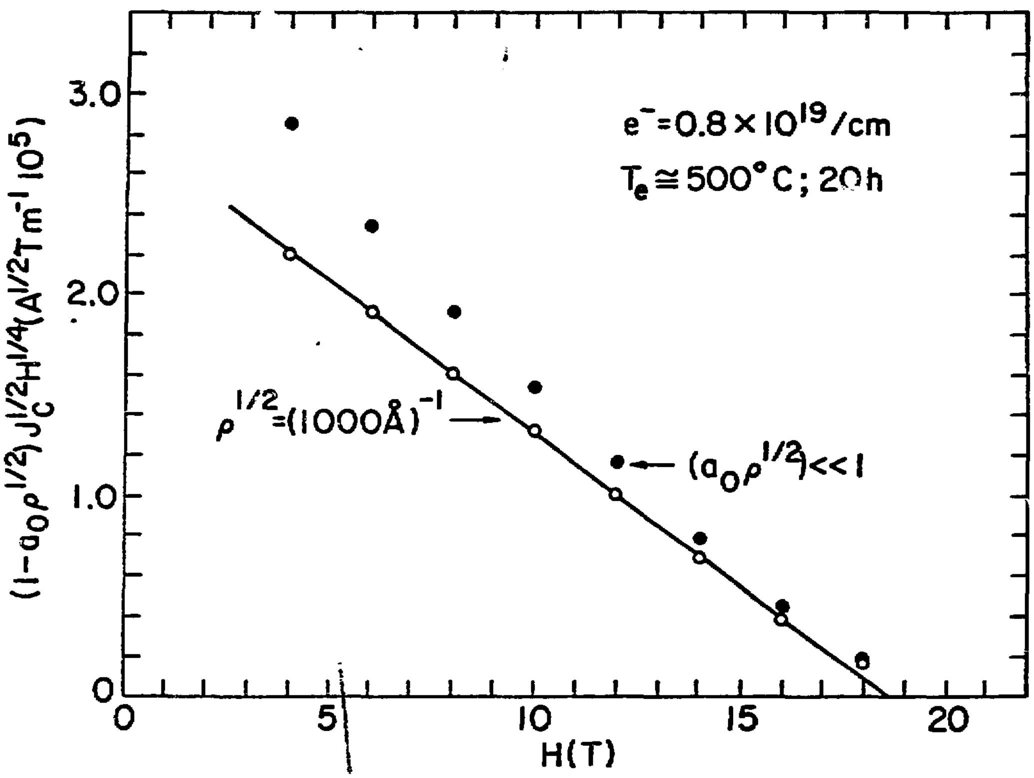

Fig. 2. A plot for $J_c^{1/2}H^{1/4}(1-a_0\sqrt{\rho})$ vs H for a Nb_3Sn wire which was electron irradiated during the heat treatment at 500°C for 24 h. This plot shows that the inclusion of $(1-a_0\sqrt{\rho})$ with $\sqrt{\rho} = (10^3\ Å)^{-1}$ is necessary to make the plot linear with H.

straight line was obtained when $1/\sqrt{\rho}$ was approximately twice as large as the grain size. This may imply that only about half of the grain boundaries are effective in pinning the flux lines. (It is well established that grain boundaries are the primary flux pinning sites in Nb_3Sn.[2])

The second category of results in which plotting $J_c^{1/2}H^{1/4}$ versus H does not yield a simple linear plot is illustrated by the behavior of bronze-processed Nb_3Sn wires which were made with a Ga-containing matrix (Cu-Sn-Ga).[10] Representative examples of $J^{1/2}H^{1/4}$ vs H plots for these wires are shown in Fig. 4. In this figure, data from Sekine and Tachikawa[11] are also included. It is speculated that the deviation from linearity for these wires is due to the paramagnetic limit on H_{c2},[10] as in V_3Ga, in which

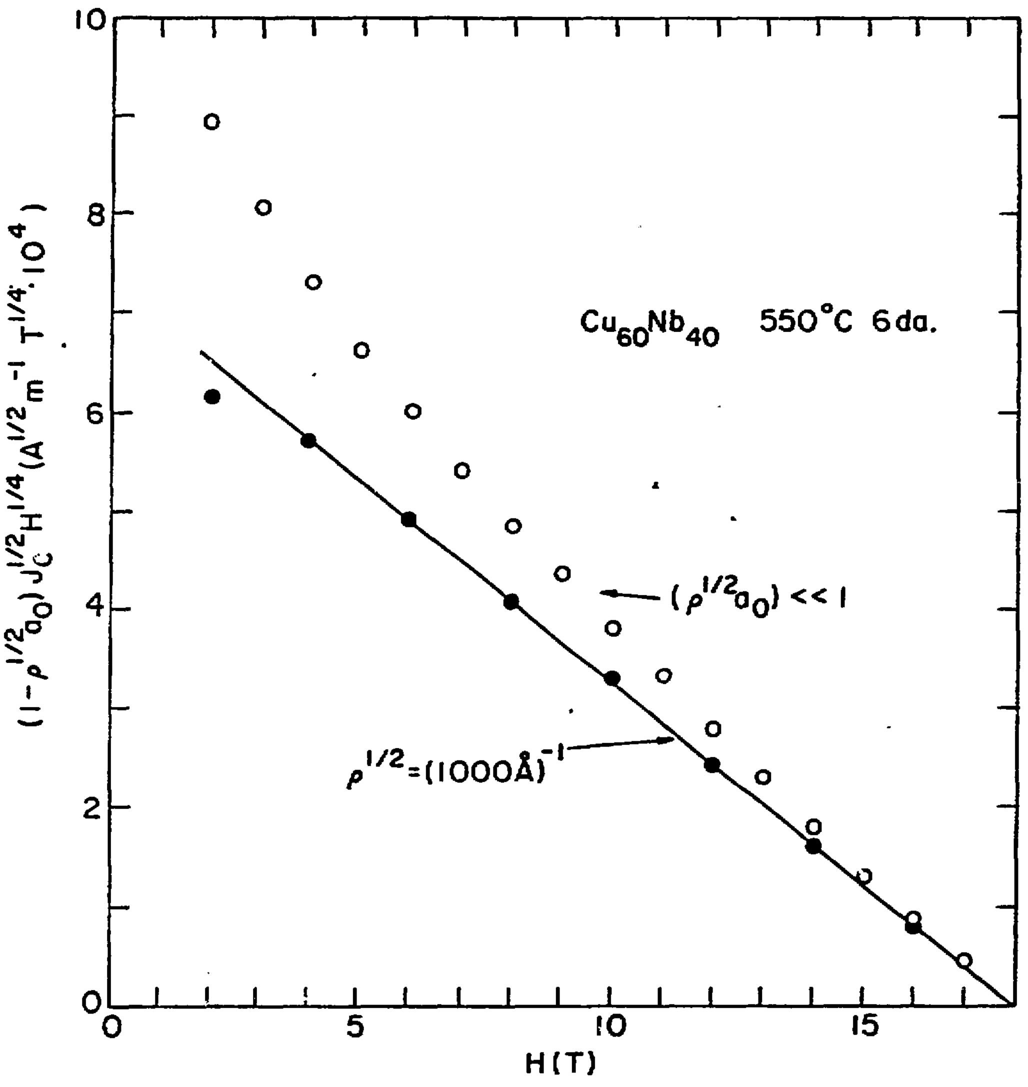

Fig. 3. A plot similar to Fig. 2 for a Nb_3Sn wire fabricated by the in-situ process [Ref. 9].

case, the conditions for which Kramer derived the scaling law do not apply.

The examples cited above, Figs. 2 and 3, appear to indicate that the Kramer scaling law adequately describes the effect on J_c of variations in the grain size of Nb_3Sn. However an experiment on the anistropy of J_c casts doubt on this conclusion. Recently, Tanaka et al.[12] reported the observation of very large differences

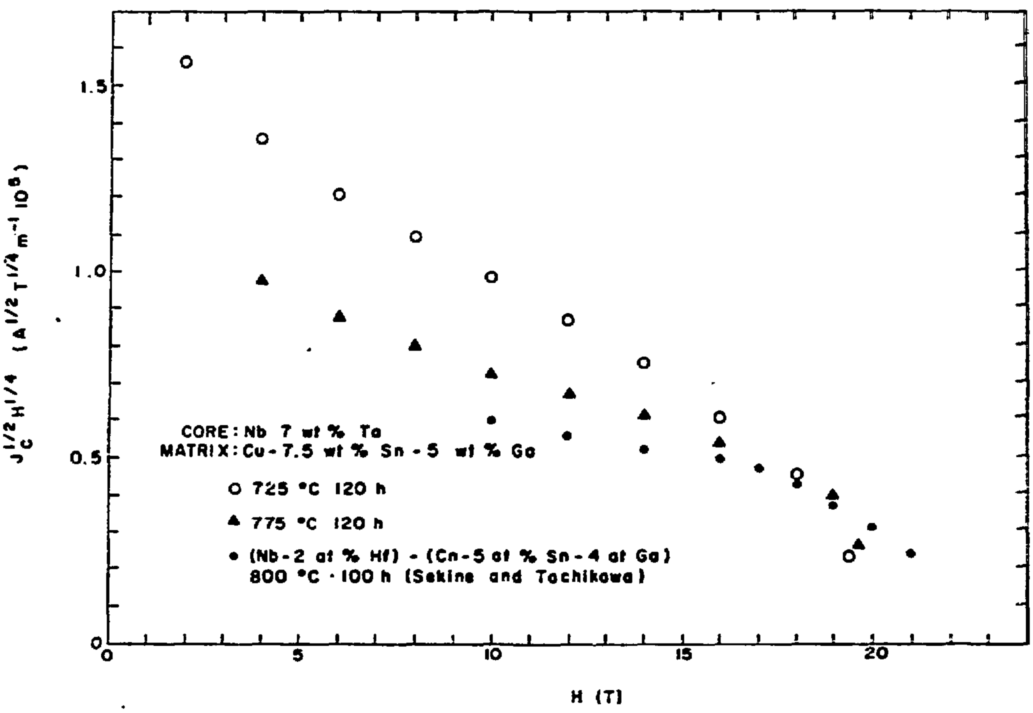

Fig. 4. $J_c^{1/2}H^{1/4}$ vs H for Ga-containing Nb_3Sn wires. These data do not appear to obey Eq. (2).

in J_c of bronze-processed V_3Ga tapes which are measured with applied fields parallel ($J_{c\parallel}$) and perpendicular ($J_{c\perp}$) to the surface of the tape. These differences are a factor of two or more, depending on the heat treatments and the value of H,[12] and they are attributed to the columnar structure of V_3Ga grains which grow perpendicular to the substrate. Thus, the effective grain size with H perpendicular to the tape is smaller than that with H parallel. The difference in J_c was accounted for by taking into account the variation in the effective grain size and assuming that J_c is inversely proportional to the grain size. The Kramer scaling law cannot be applied to these results because of paramagnetic limitation in V_3Ga. However such an experiment for Nb_3Sn is revealing.

We have performed a similar experiment for Nb_3Sn using a flattened wire which had a matrix-to-core ratio of $\sim$15. Pieces of the wire were heated for 32 h at 725°C and 4 h at 775°C. In both specimens, the $J_c^{1/2}H^{1/4}$ vs H plots are straight in the field range of $\sim$5 to $\sim$10 tesla regardless of the orientation of H to the tape surface, as shown in Fig. 5. As found by Tanaka et al.,

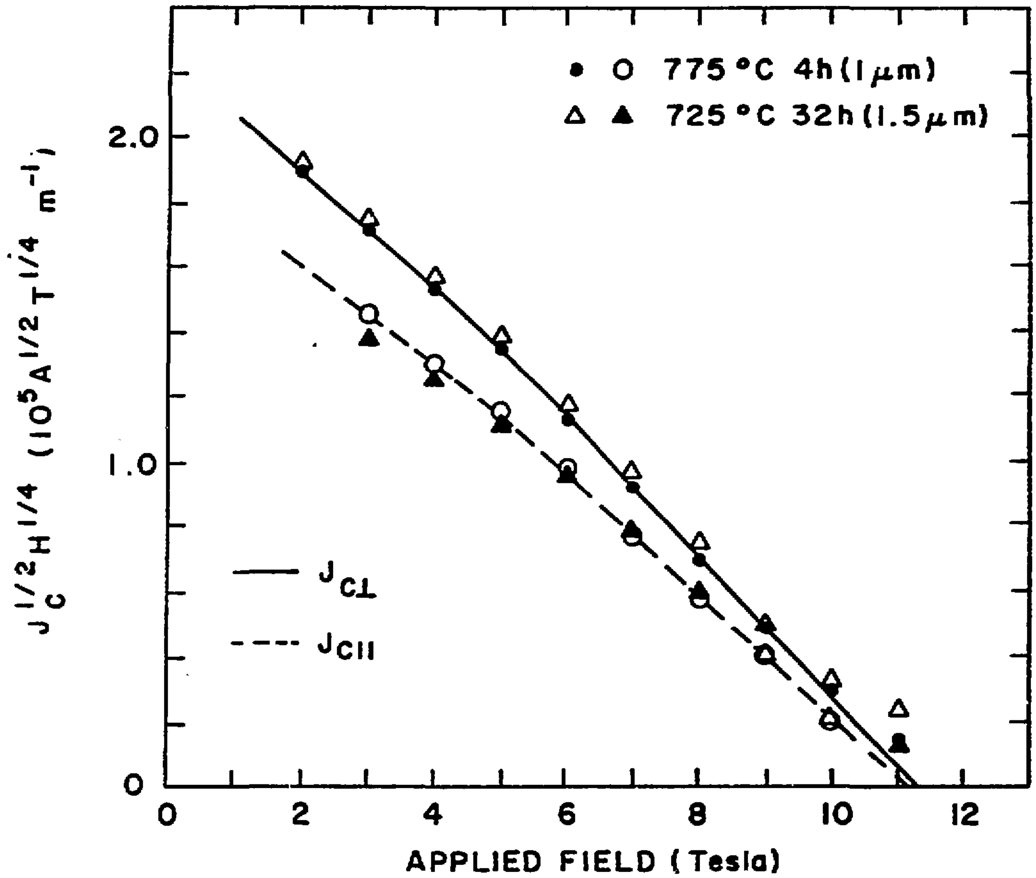

Fig. 5. Plots for $J_c^{1/2}H^{1/4}$ vs H for H parallel ($J_{c\parallel}$) and perpendicular ($J_{c\perp}$) to the surface of Nb_3Sn tapes.

$J_{c\perp}$ is larger than $J_{c\parallel}$, but only by 30 to 40%. These smaller differences in J_c are due to nearly equiaxed growth of Nb_3Sn grains in these specimens, as observed with a scanning electron microscope. However, these results clearly reveal a difficulty with Eq. (2) as a description of the grain-size dependence of J_c. Variations of J_c by as much as 30-40% caused by anisotropy in the grain size imply, in Eq. (2), that the $a_o\sqrt{\rho}$ term must be large enough that a plot of $J_c^{1/2}H^{1/4}$ vs H would exhibit an easily observable concave-up curvature, as in Figs. 2 and 3. Yet, as seen in Fig. 5, the results show no such curvature and imply that $a_o\sqrt{\rho} << 1$, in which case variations in $a_o\sqrt{\rho}$ would cause a negligible variation in J_c. Clearly there is a contradiction here, and its resolution is not obvious at present. Anistropy in H_{c2} (and thus κ_1) with crystallographic orientation is not likely to be the origin of anisotropy in J_c, since, as seen in Fig. 5, the extrapolated H_{c2} from $J_{c\perp}$ and $J_{c\parallel}$ is essentially the same in both cases. Unfortunately, the values of H_{c2} for these tapes are significantly lower than those for Nb_3Sn wires with the same matrix-to-core ratio. The cause for the reduction of H_{c2} is not understood at this time. However, we believe that the observed differences in $J_{c\perp}$ and $J_{c\parallel}$ and our conclusion about the scaling law drawn from the present data are valid in spite of the unexplained reduction in H_{c2} of these specimens.

As a further test of the scaling law, the behavior of the values of κ_1, as determined by Eq. (2) from the slope of $J_c^{1/2}H^{1/4}$ versus H plot, were studied for a set of monofilamentary wires which are heat treated for 6, 16, and 64 h at 725°C and 96 h at 675°C. The ratio of bronze-to-core for these wires was ~7.6. The data are plotted in Fig. 6 as $J_c^{1/2}H^{1/4}$ vs H, and the lack of concave-up curvature over the field range 8-15 tesla implies that $a_o\sqrt{\rho} \ll 1$. Values of κ_1 obtained from the slopes of these plots, as suggested by Eq. (2), are listed in Table 1. (The data from this set of specimens was chosen as an illustration, but essentially identical behavior was observed for wires with other bronze-to-core ratios.) The values of κ_1 so obtained are smaller than that expected for Nb_3Sn, for which the smallest expected value is ~20 for very clean specimens.[13] A quantitative discrepancy of this order is perhaps not unreasonable considering the uncertainty in the value of the shear constant, C_{66}, for the flux line lattice which was used in the derivation of Eq. (1). What is a more serious difficulty is the fact that the observed value of the κ_1 increases with heat treatment time while the observed value of (H_{c2}/T_c) is unchanged (see Table 1). This difficulty is best illustrated by examining the relationship between (H_{c2}/T_c) and κ_1[3,13,14]:

$$\frac{H_{c2}}{T_c} = c[N(0)(1+\lambda)]^{1/2}\,(1-t^2)\kappa_1 = c'\gamma^{1/2}(1-t^2)\kappa_1 \qquad (3)$$

where c and c' are numerical constants, N(0) is the density of states at the Fermi level, λ is the electron-phonon coupling constant, γ is the electronic specific heat coefficient and t = $= T/T_c$. Recent experimental studies[14-17] of the effect of disorder and nonstoichiometry on the superconducting properties of a variety of A15 compounds show N(0), λ, and γ to be monotonically increasing functions of T_c. Our data, Table 1, clearly show that T_c increases by ~4% on increasing the reaction time from 6 h to 64 h at 725°C. Thus one would expect, on the basis of the experimental results cited above, that N(0), λ, etc. would increase by roughly the same amount. Since our data show H_{c2}/T_c to increase by about 3%, one would then expect κ_1 to remain more-or-less constant, yet, as seen from Table 1, the value of κ_1 deduced with

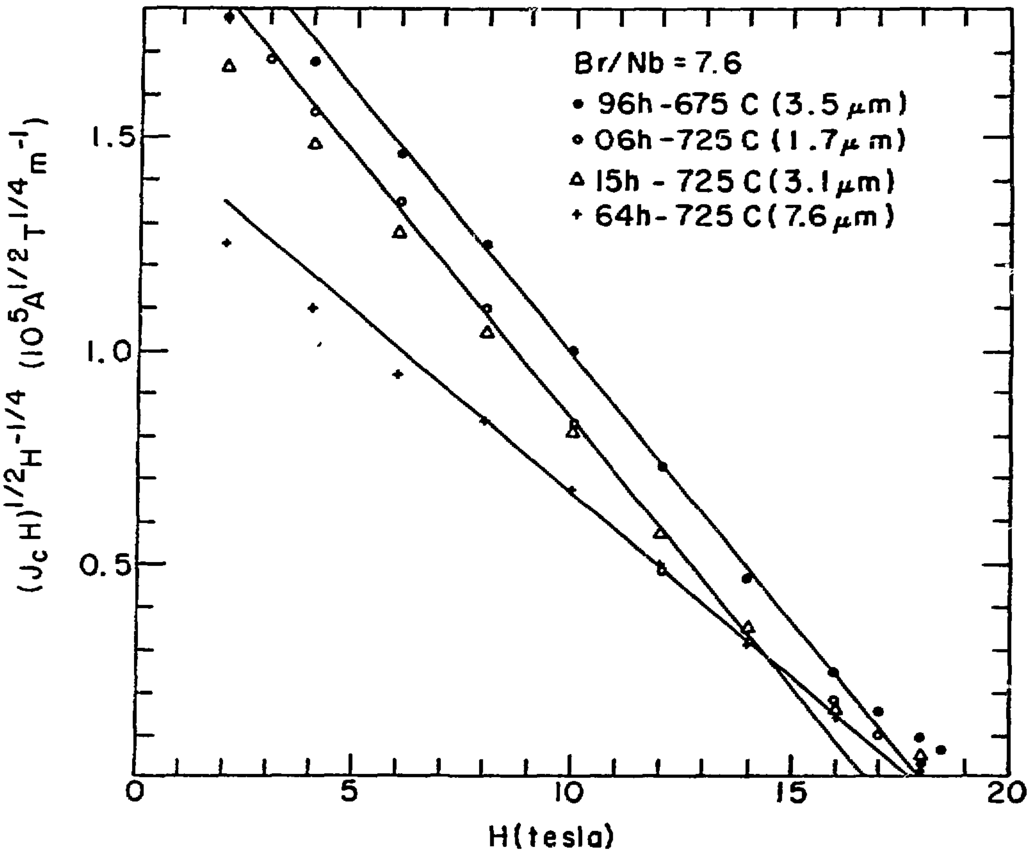

Fig. 6. Plots for $J_c^{1/2}H^{1/4}$ vs H for a series of Nb_3Sn wires which were heat treated for 6, 16, and 64 h at 725°C and for 96 h at 675°C.

the scaling law increases by ∿50%! This discrepancy suggests that even though the <u>form</u> of the scaling law for flux pinning at high fields derived by Kramer seems to be obeyed for Nb_3Sn, the interpretation of parameters such as "κ_1" which appears in it may not be the same as assumed in Kramer's derivation.

CONCLUSION

The scaling law derived by Kramer for magnetic flux pinning in high magnetic fields was examined for its applicability to the magnetic field dependence of critical-current densities in the bronze processed monofilamentary Nb_3Sn wires. From this we

Table 1. Superconducting Properties of the Nb_3Sn Wires

	6h/725°C	16h/725°C	64h/725°C	96h/675°C
κ_1	9.2	9.9	13.8	8.66
$T_c^{(a)}$	16.15	16.45	16.85	16.80
$H_{c2}^{(b)}$	16.6	16.9	17.5	17.6

[a]The midpoint T_c measured inductively with the bronze matrix still present.

[b]Determined by the linear extrapolation according to Eq. (2).

conclude about the scaling law that: 1) its prediction for the form of the dependence of critical current on magnetic field and grain size $\{|\bar{J}_c \times \bar{H}| \sim h^{1/2}(1-h)^2(1-a_o\sqrt{\rho})^{-2}\}$ was found to be very good in most cases including wires with very small Nb_3Sn grains (∿400 Å). It was found very useful in comparison of J_c for different wires and in extrapolating to obtain H_{c2} for these wires. 2) However, it could not account consistently for the anisotropy in critical current of a tape which was measured with H applied perpendicular and parallel to the tape face. 3) The values of κ_1 which were determined with the scaling law were too small by a factor of 2 to 3, and the trend in the variation with heat-treating time was opposite to that which is reasonably to be expected. That the behavior of κ_1 is thus seriously in contradiction with the expected behavior for Nb_3Sn suggests basic faults in the derivation of the scaling equation for critical currents at high magnetic fields.

ACKNOWLEDGMENTS

The authors appreciate technical assistance by A. Cendrowski, D. Horne, R. Mitchell, F. Perez, and R. Sabatini and the use of the high magnetic field facilities at the Francis Bitter National Magnet Laboratory.

REFERENCES

1. For a review see M. Suenaga, W. B. Sampson, and C. J. Klamut, IEEE Trans. on Magnetics MAG-11:231 (1975).
2. For a review see, J. D. Livingston, Kristal und Technik 13: 1379 (1978).
3. For a review see, D. O. Welch, to be published in Adv. Cryo. Engin. 25 (1980).
4. E. J. Kramer, J. Appl. Phys. 44:1360 (1973).
5. For example, D. U. Gubser, T. L. Francavilla, D. G. Howe, R. A. Muessner, and F. T. Ormand, IEEE Trans. on Magnetics MAG-15:385 (1979); T. S. Luhman, C. S. Pande, and D. Dew-Hughes, J. Appl. Phys. 47:1459 (1976).
6. G. Rupp, E. Springer, and S. Roth, Cryogenics, 17:141 (1977).
7. M. Suenaga, T. Onishi, D. O. Welch, and T. S. Luhman, Bull. Am. Phys. Soc. 23:229 (1978), and unpublished data.
8. C. L. Snead, Jr. and M. Suenaga, Appl. Phys. Lett. 36:474 (1980).
9. D. K. Finnemore and J. D. Verhoeven, to be published in Prog. in Cryo. Engin. 25 (1980).
10. C. L. Snead, Jr. and M. Suenaga, IEEE Trans. Magnetics MAG-15:625 (1979).
11. H. Sekine and K. Tachikawa, Appl. Phys. Lett. 35:472 (1979).
12. Y. Tanaka, K. Itoh, and K. Tachikawa, J. Japan Inst. of Metals 40:515 (1977).
13. T. P. Orlando, E. J. McNiff, Jr., S. Foner, and M. R. Beasley, Phys. Rev. B 19:4545 (1979).
14. H. Wiesmann, M. Gurvitch, A. K. Ghosh, H. Lutz, O. F. Kammerer, and M. Strongin, Phys. Rev. B 17:122 (1978).
15. A. K. Ghosh and M. Strongin, Proc. of the 1979 Conf. on Superconductivity in d- and f-Band Metals, LaJolla, CA (in press).
16. F. Y. Fradin and J. D. Williamson, Phys. Rev. B 10:2803 (1971).
17. R. Viswanathan and R. Caton, Phys. Rev. B 18:15 (1978).