HIGH TEMPERATURE SUPERCONDUCTORS: THE CURRENT STORY

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ABSTRACT

The problem of critical current density J_c of high temperature superconductors forms the theme of the 'current' story.

INTRODUCTION

ALTHOUGH during the last 12 months La–Sr–Cu–O and Y–Ba–Cu–O and their related other systems possessing the critical temperature T_c in the range of 30 K to 95 K have been extensively studied, the problem of increasing the critical current density J_c remains one of the main challenges of high temperature superconductivity research today. These materials are extreme type-II superconductors having the Ginzburg-Landau parameter k in the range of 70 to 100. There is a considerable scatter in the reported data[1–3] and some of the parameters for polycrystalline samples are given in table 1.

The type-II behaviour of the high T_c ceramic materials is however complicated by the two factors: (i) anisotropy effects, and (ii) the granular nature and the presence of weak links. These aspects also hold for the two recently discovered Bi and Tl based high T_c oxides containing Sr, Ca and Cu. Both the factors severely restrict the overall current-carrying capacity of high T_c ceramic materials.

The values of the lower critical field $B_{c_1}(O)$ (table 1) experimentally determined from the point of deviation of the magnetization from the Meissner effect are found to be consistent with the GLAG relation

$$B_{c_1}(O) = [B_c(O)/\sqrt{2}k] \ln k \qquad (1)$$

as well as with the values estimated from the parameters γ, ρ_n and T_c, using the expression given by Echarri *et al*[4]

$$B_{c_1}(O) = (150.5T_c/\rho_n) \ln (1.152 \times 10^{-2} \rho_n \gamma^{1/2}). \qquad (2)$$

The anisotropy effects are linked with the unusual structural features, namely an orthorhombic unit cell containing highly conducting Cu–O planar networks in the a–b plane. Studies on single crystals have revealed considerable anisotropies in $B_{c_1}(O)$, $B_{c_2}(O)$, and consequently also with respect to $\xi(O)$, when the parameters are measured parallel and perpendicular to the a–b plane. Typically for $YBa_2Cu_3O^5_{9-y}$ (i.e. the 123 compound) $B_{c_2}(O)_{\parallel} \simeq 200$ T, $B_{c_2}(O)_{\perp} \simeq 25$ T, $\xi(O)_{\parallel} \simeq 35$ Å and $\xi(O)_{\perp} \simeq 6$ Å while the same parameters for $Bi_2Sr_2CaCu_2O^6_X$ have been found to be around 400 T, 21 T, 40 Å and 2.5 Å respectively. These anisotropies make the behaviour of polycrystalline samples texture sensitive and dependent on processing conditions.

The polycrystalline-sintered compacts consist of superconducting grains that are mutually weakly coupled which results in the manifestation of Josephson tunnelling effects in bulk samples[7]. Because of very small coherence length normal to the a–b plane the tunnelling of electron pairs across the grain (or twin) boundaries can become important even in thin films deposited under clean conditions[8]. The presence of weak links is found to cause departure from the ideal type-II behaviour by a small flux slippage occuring at a field $B^*_{c_1} \ll B_{c_1}$ which is followed by a low magnetic hysteresis. $B^*_{c_1}$, which is less than 5 Oe[9] at 77 K for 123 compounds, is the lower critical field of the weak links and not of the superconducting grains as mistakenly assumed recently by Grover *et al*[10].

The large thermodynamic critical field $B_c(O)$ accompanied by a modest value of the penetra-

Table 1 *Parameters of the two oxides* $La_{1.85}Sr_{0.15}CuO_{4-y}$ $YBa_2Cu_3O_{9-x}$

ξ (0)	25 Å	12 Å
λ (0)	2500 Å	1400 Å
B_c (0)	4.5–6 kOe (0.45–0.6 T)	8–13 kOe (0.8–1.3 T)
B_{c2} (0)	650 kOe (65 T)	1000–1800 kOe (100–180 T)
B_{c1} (0)	200–300 Oe	250–40 Oe

tion depth $\lambda(0)$ lead to a significantly high depairing current density J_d which is about 5×10^8 A/cm^2 for the 123 compound. But this aspect or B_{c_2} alone does not render the material an ability to sustain a large J_c in a transverse high magnetic field. Above B_{c_1} the material enters the mixed state, comprising of a triangular lattice of Abrikosov's flux lines (FLL). Each flux line has a normal core of radius ξ carrying a quantum of magnetic flux $\phi_0 = 2 \times 10^{-7}$ G.cm^2 $(2 \times 10^{-15}$ Wb) and is surrounded by vortices of supercurrents spread over a length λ. The transport current in the mixed state imposes the Lorentz driving force $\bar{F}_L = \bar{B} \times \bar{J}$ per unit volume on the FLL. In a static non-uniform configuration of curved flux lines the driving force per unit volume is given by[11] $F_L = (\bar{B}/4\pi)X$ curl $\bar{H}(\bar{B})$ which, for the situation of two-dimensional system of straight flux lines, reduces to $F_L = (\bar{B}/4\pi)\mathrm{d}\bar{H}(\bar{B})/\mathrm{d}\bar{x}$, where $\bar{H}(\bar{B})$ is the external field in equilibrium with the local induction $\bar{B}$. If the flux lines move a voltage is developed and the current ceases to be lossless. The way to circumvent this is to pin the FLL and thereby prevent the flux motion. Inhomogeneities present in the material offer pinning forces F_p counteracting the driving force whereby a static non-uniform flux distribution becomes permissible, provided $F_L < F_p$. The critical current is attained when the driving force is just equal to the pinning force, i.e., $\bar{B} \times \bar{J}_c = \bar{F}_p$. When the force balance condition is fulfilled the sample attains the 'critical state' where the field gradient $\mathrm{d}\bar{H}(\bar{B})/\mathrm{d}\bar{x}$ is maximum or critical and the sample carries a critical current density J_c which is a function of the local value of the flux density B. The advantage of this concept[12,13] is that if the relation between the critical current and the field is known, the field profile within the superconductor can be determined and the magnetization curve can be calculated. Alternatively, from the measured magnetization data $J_c(B)$ of the material can be deduced. Although for mere simplicity Bean had originally considered the critical state equation where J_c was independent of B the concept is fairly general to take care of the real situation in the range where B is larger than the field corresponding to the peak magnetization. This approach has worked extremely well with the conventional superconductors, and $J_c(B)$ estimated from magnetization curves is in good accord[14,15] with the transport current measurements. In the case of ceramic superconductors, however, making of satisfactory lead connections has been a problem and thus magnetization measurements have been relied upon for determining $J_c(B)$.

Flux pinning by inhomogeneities arises because the latter give rise to a local variation in the superconducting properties where the FLL energy is locally reduced to get pinned. The flux line energy has two main components, one due to its normal core and the other due to the circulating currents. The two components respectively give rise to the core interaction and the magnetic interaction. In large k materials the magnetic interaction is considered to be weak. The ideal situation for optimum core interaction is when the pinning

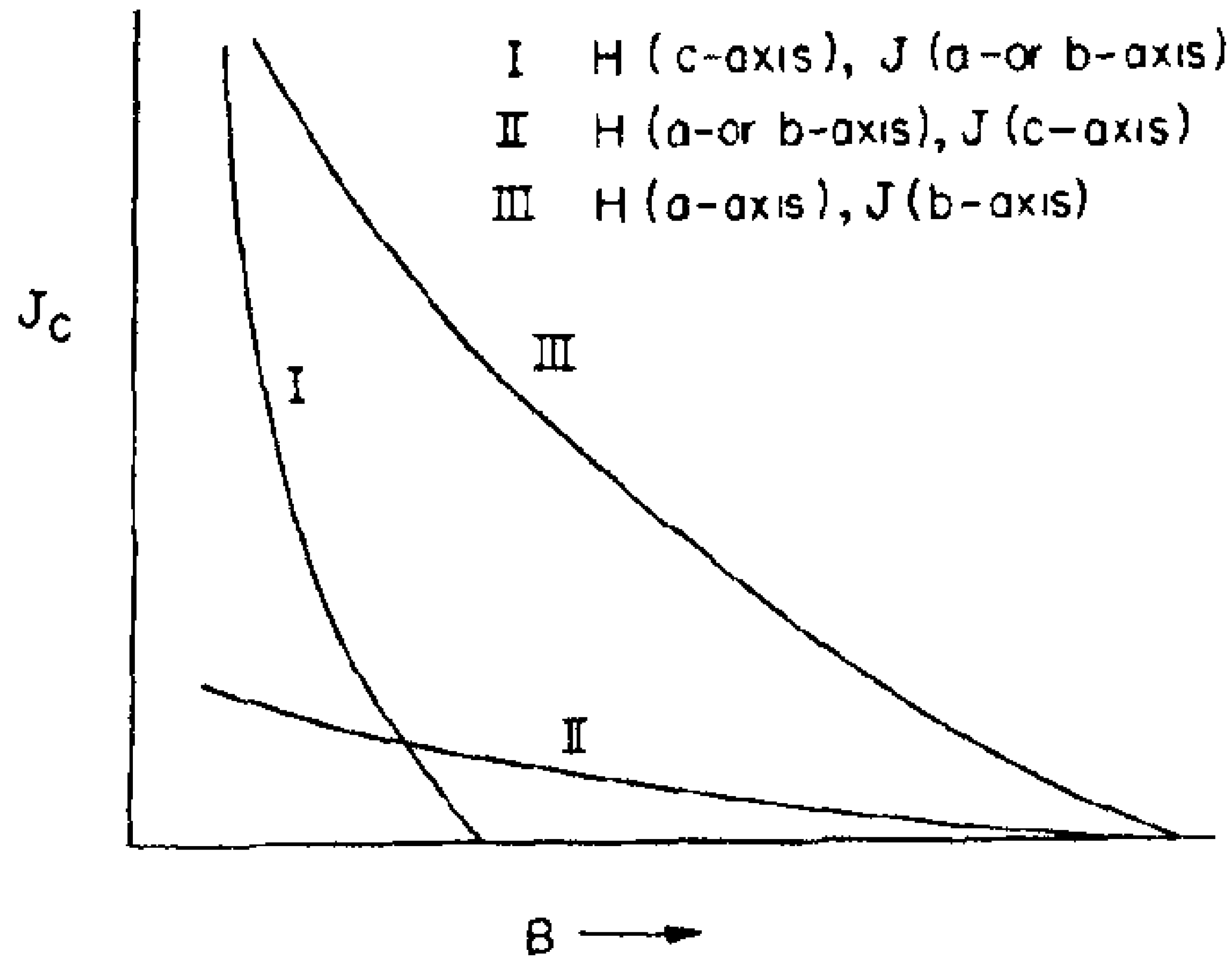

Figure 1. $J_c(B)$, three orientations.

centres are normal particles of radius ξ. The core energy per unit length of the flux line is approximately $B_c^2\xi^2/8$. To displace the core by a distance ξ would require a force of $B_c^2\xi/8$ and the corresponding current density would be $[B_c^2\xi/8\phi_0](1-b)$, where $b = B/B_{c_2}$ and the factor $(1-b)$ represents some arbitrary field dependence of J_c. In the case of single crystal of high T_c ceramic superconductor, e.g. 123 compound, one can predict J_c for three different orientations of mutually perpendicular field and current:

(I) H (*c*-axis):J_c (*a* or *b*-axis) $= [B_c^2\xi_{\parallel}/8\phi_0](1-b)$,

(II) H (*a* or *b*-axis):J_c (*c*-axis) $= [B_c^2\xi_{\perp}/8\phi_0](1-b)$,

(III) H (*a*-axis):J_c (*b*-axis) $= [B_c^2\xi_{\parallel}/8\phi_0](1-b)$.

In the situation (I), as described earlier, B_{c_2} has the lowest value. J_c curves for three orientations are depicted in figure 1. As compared to the orientation (III), both (I) and (II) are unfavourable for high $J_c(B)$.

From the view point of flux motion also, orientation (III) is likely to offer constraints and J_c is thus expected to be higher. The motion of FLL is believed to take place through the movement of dislocations in FLL. Because of very small range of coherence in the *c*-direction, comparable to the lattice spacing, a force equivalent to the Peierls–Nabarro force, which is known to oppose the movement of crystal dislocations, is expected to be dominant in FLL which would inhibit the easy motion of flux line dislocations.

Thus, in polycrystalline samples J_c gets significantly lowered because of the presence of

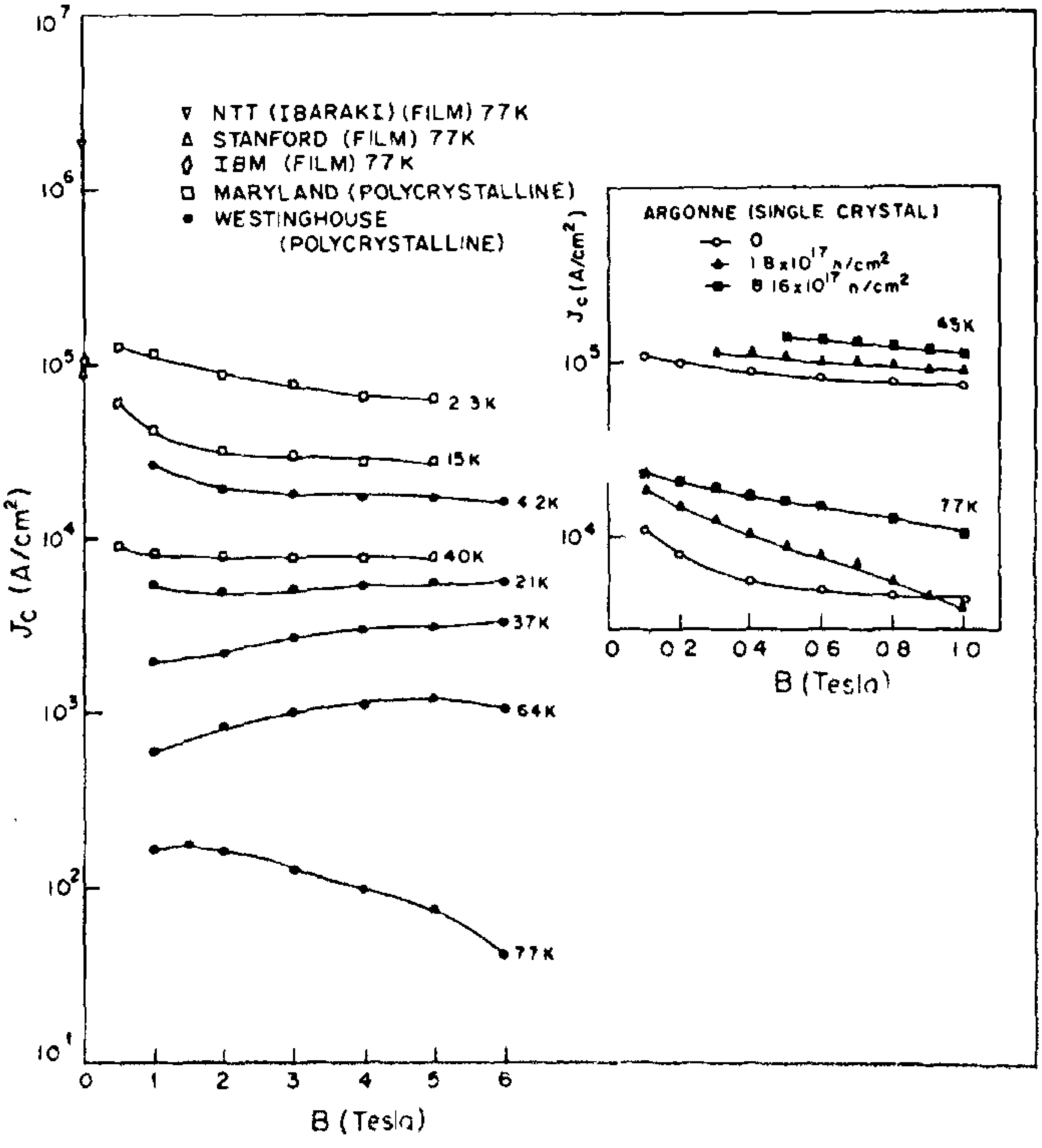

Figure 2. $J_c(B)$ data at different T.

unfavourably oriented grains. Efforts are currently being directed to produce wires and tapes having properly oriented grains for optimum $J_c(B)$.

It would be worth examining some of the reported $J_c(B)$ data for 123 compound in the form of (i) polycrystalline sintered compacts[16,17], (ii) bulk single crytals, and (iii) deposited thin films[5,18–21] (figures 2 and 3). Low field J_c values for thin films are about three orders of magnitude larger than for polycrystalline materials. The material sintered and heated above its melting temperature at Bell Labs yielded[22] the zero field J_c of 7.4×10^3 A/cm^2 at 77 K (figure 3) and more recently this value has been raised to 1.7×10^4 A/cm^2. The bulk single crystals are found to possess J_c values intermediate between thin films and polycrystalline sintered compacts. In figure 4 we have drawn $F_p(b)$ curves at different temperatures for the data of figure 2 for the sintered samples. Some of the general features observed may be summarized as follows:

[1] The general scaling law for the volume pinning force, given by[11]

$$F_p(b, T) \propto B_{c_2}(T)^m b^n (1-b)^l \qquad (3)$$

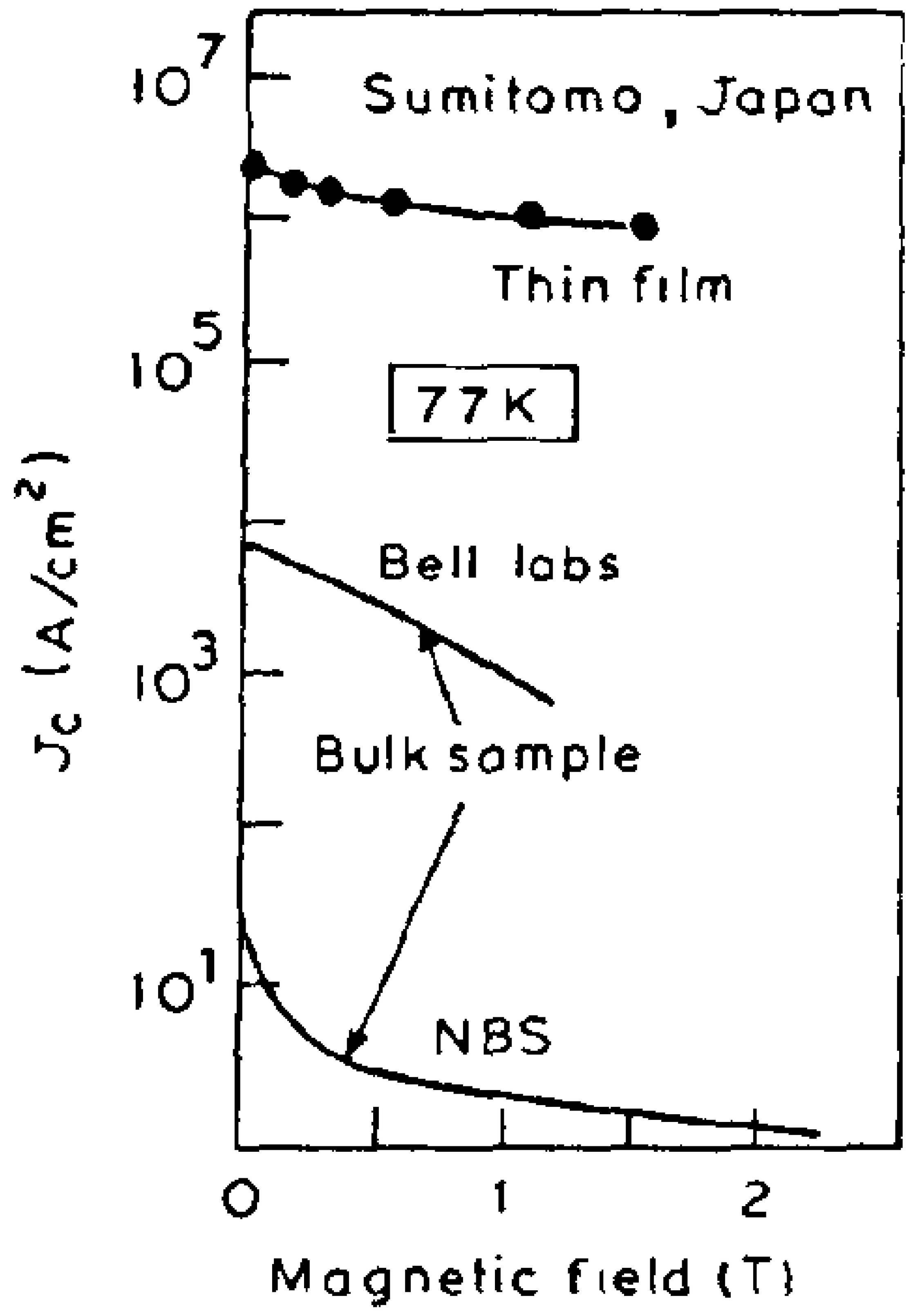

Figure 3. $J_c(B)$ data at 77 K.

where $b = B/B_{c_2}(T)$ and $m \approx 2.5$, $n \approx 0.5$ and $l \approx 2$ does not seem to hold. Decrease of J_c with T is much stronger for polycrystalline bulk samples than for the usual type-II superconductor. The exponent m is much larger, i.e. 4 to 6 instead of 2 or 2.5 found for the conventional superconductors. Single crystals have m 2.5.

[2] In the measured field interval up to 5 T or more, contrary to the decrease exhibited by the conventional superconductors, J_c either remains constant or even slightly increases with field.

[3] The $F_p(B)$ curves for bulk single crystals show the existence of maxima in F_p at very low values of b_m less than 0.02 or they give indications of rather broad maxima in polycrystalline samples. The maxima tend to shift to a higher b_m value as the temperature is lowered (figure 5).

[4] $J_c(B)$ values for powdered samples, as estimated from magnetization data, are about two orders of magnitude larger than for bulk samples before they were crushed.

There are instances where J_c of both thin films and bulk single crystals[24,25] of 123 compound, particularly at low fields, decreases exponentially with B. But there is nothing particularly unusual about this. The critical state equation is determined solely by the microstructure and, in principle, $J_c(B)$ can have any form and the same sample may show different field dependences of J_c in different range of B values[26,27] (figure 8). In fact, the exponential field dependence of J_c has been observed in the conventional superconductors like Nb–Zr alloys (figure 6)[28] and superfine filaments of Nb–Ti (figure 7), the latter exhibiting a dominant surface pinning. Also, bulk Nb–Ti[26] and Nb–Zr–Ti[27] alloys (figure 8) as well as annealed Nb–Ta[29] (inset of figure 8) in the low field range show exponential field dependence of J_c. There has been a suggestion[30] that the exponential field dependence of J_c in ceramic superconductors might be due to a strongly field dependent electron pairing distinctly different from the phonon-mediated

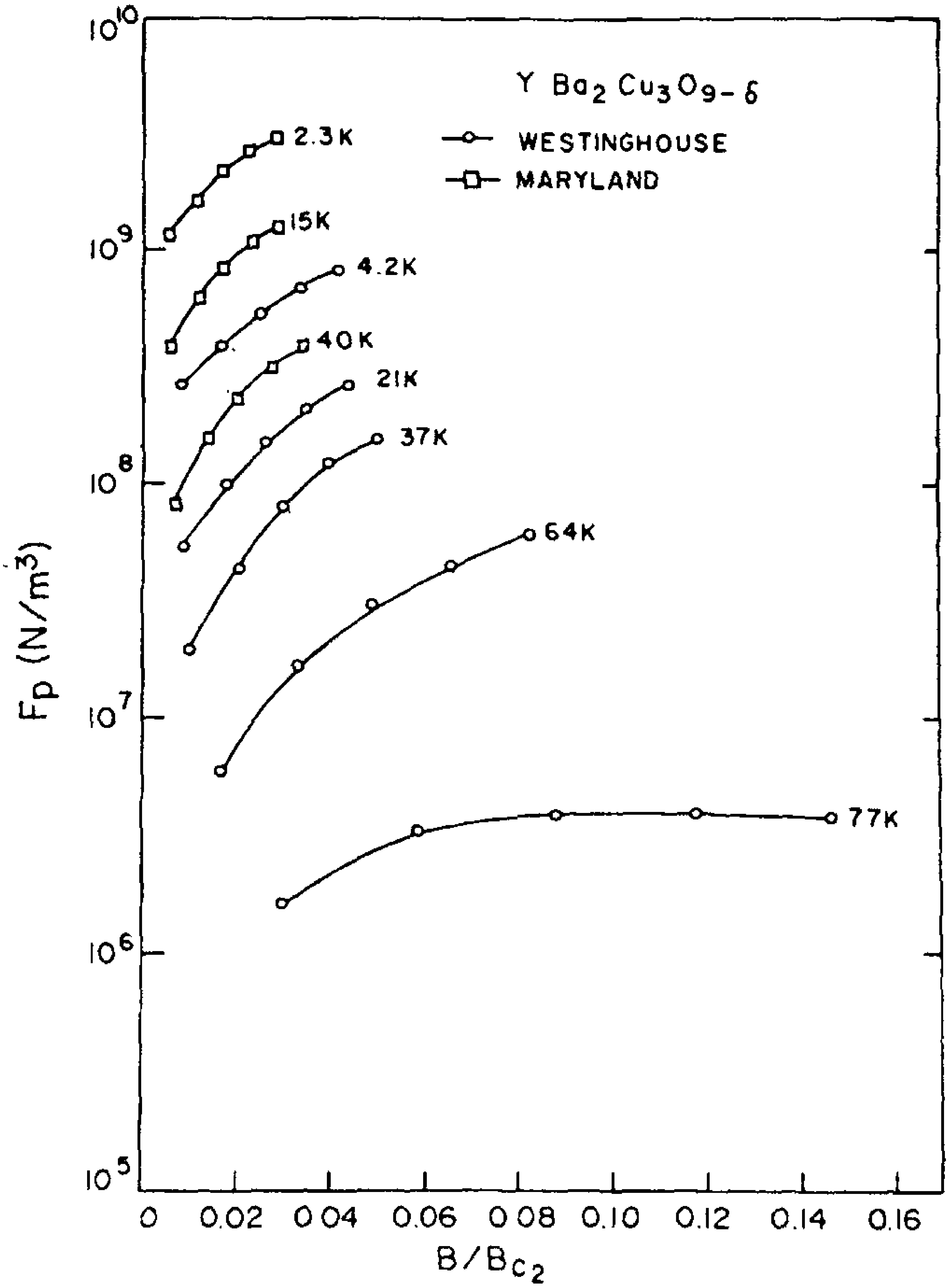

Figure 4. $F_p(B)$ curves for sintered compacts.

BCS type and it is argued that this would also explain the observed positive curvature in B_{c_2} vs T curves close to T_c found for the ceramic superconductors. In the above background this seems unnecessary and possibly also incorrect. Moreover, the positive curvature of B_{c_2} vs T curves has been found also for some of the conventional layered superconductors such as $NbSe_2$[31] and TaS_2[32] which are known to exhibit anisotropy effects. Both anisotropy and Josephson coupling of grains have previously been advanced to explain the unusual features of B_{c_2} vs T curves found in the conventional superconductors[33,34]. Similar explanation should hold for ceramic superconductors known to be anisotropic and Josephson coupled. Thus, it seems more logical to relate the $J_c(B)$ data for ceramic superconductors to microstructural features, surface pinning effects and the presence of weak links rather than to anything else.

At T close to T_c, the current is essentially Josephson weak-link like, decreasing rapidly with increasing B. In single crystals of poor quality the twin boundaries can serve as weak links and essentially the same effect may result. In suitably oriented films containing Ho (Sumitomo, Japan, figure 3)[21], where J_c is high

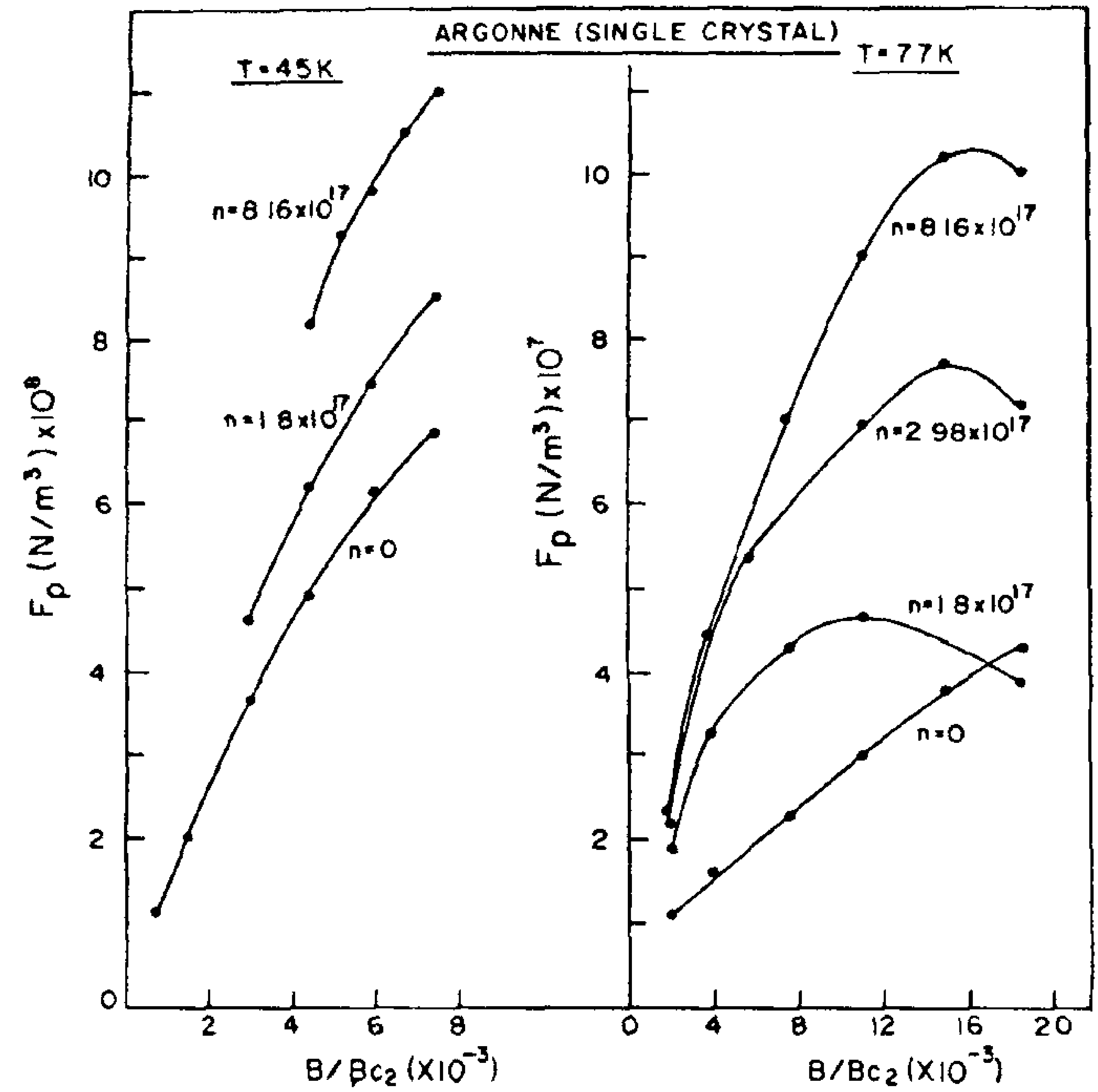

Figure 5. $F_p(B)$ curves for irradiated single crystals.

and is much less sensitive to B, the effect can be understood in terms of a stronger flux pinning resulting from a more favoured orientation (figure 1). Also, as the current flows in the a–b plane having a larger range of coherence, the adverse effect of weak links is expected to be less.

As the temperature is lowered the grains get coupled more strongly owing to enhanced proximity effect and the interconnections become fully superconducting with reduced order parameter. These regions can provide additional pinning sites at low temperatures. Detailed calculations have been made by Kramer and Freyhardt[35] for the proximity effect pinning for the above configuration, but in the 'clean limit' for the normal regions.

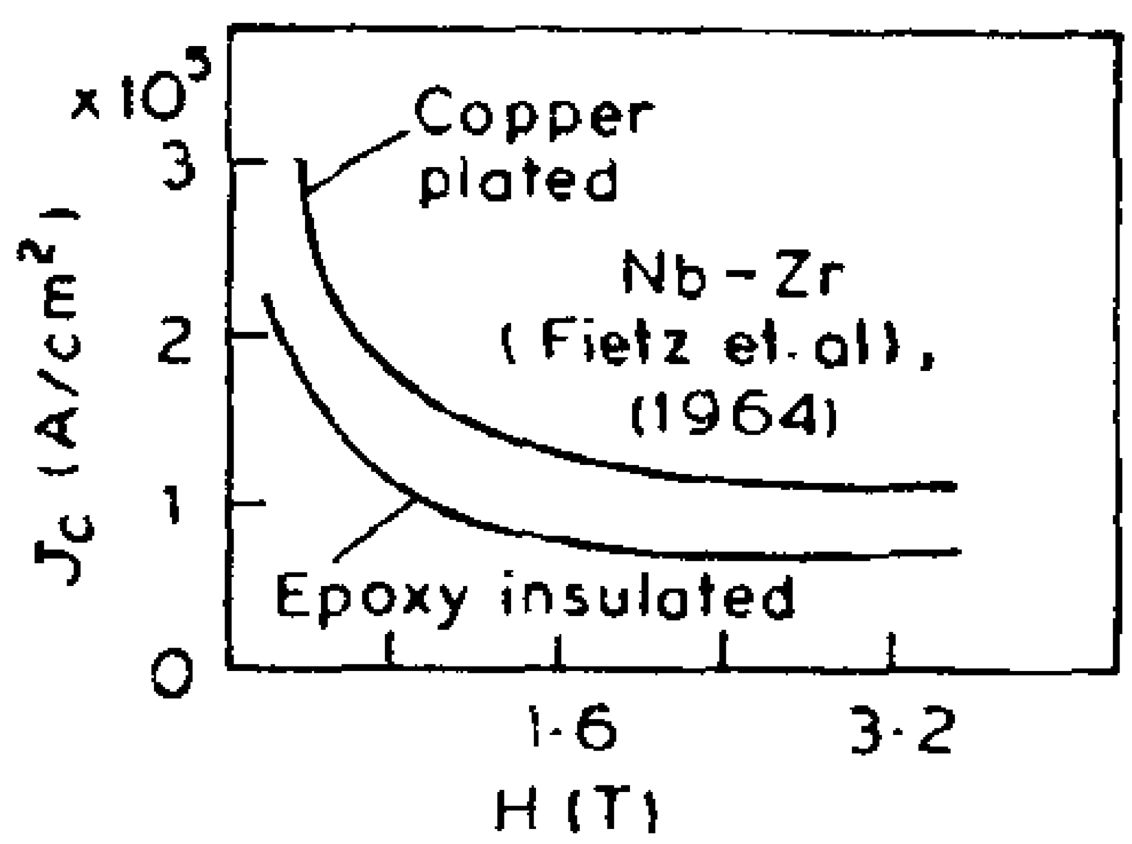

Figure 6. $J_c(B)$, exponential dependence for Nb-Zr samples.

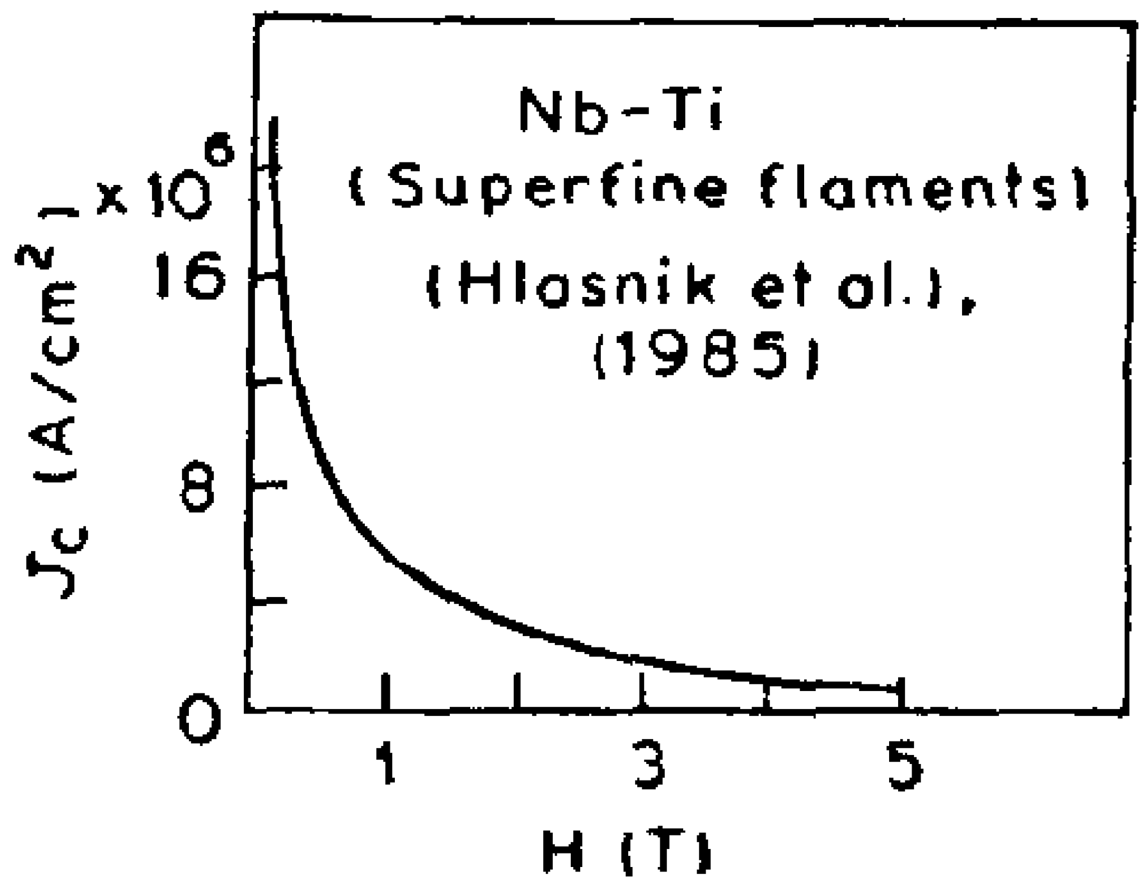

Figure 7. $J_c(B)$, exponential dependence for Nb-Ti (superfine filaments).

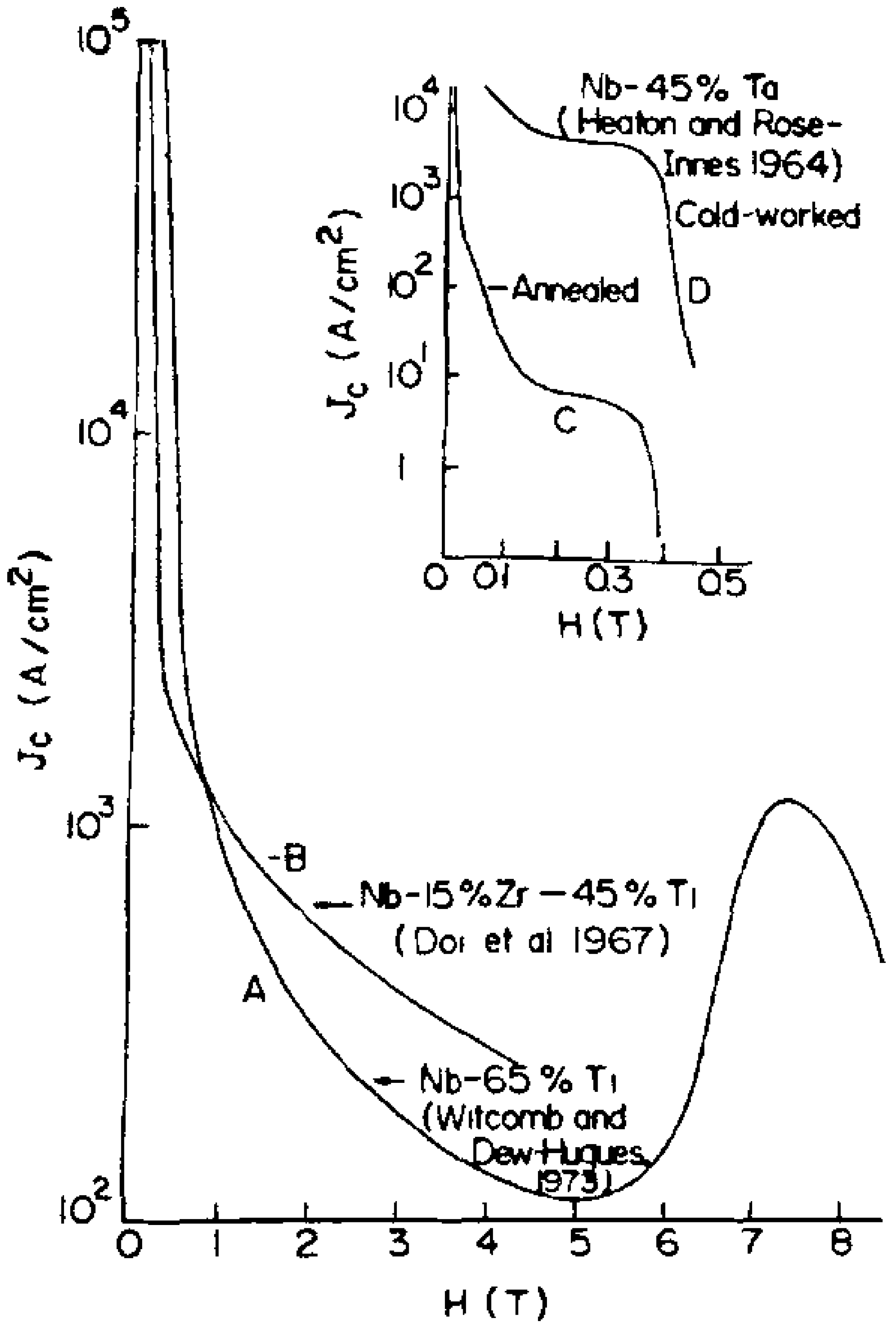

Figure 8. Exponential dependence of J_c on B for Nb-Ti and Nb-Zr-Ti alloys in low fields. Inset: low field exponential dependence for Nb-Ta alloy.

Their results show the exponent m to increase from 2.5 to 3.5. Narasimha Rao *et al*[36] recently examined the situation in the 'dirty limit' where the order parameter is found to be[37]

$$\Delta = \frac{\cosh(px/\xi_n)}{\cosh(pd_n/2\xi_n)}, \quad (4)$$

where $p^2 = 1 + (2\pi B\xi_n/\phi_0^2)\lambda^2$, d_n = thickness of the interconnection and ξ_n (in the dirty limit) = $(\hbar V_F/6KT)^{1/2}$. The ratio of the order parameter minimum to its value at the interface is equal to

$$Q = 1/\cosh(pd_n/2\xi_n). \quad (5)$$

Due to the nearly exponential dependence in some temperature range where the argument of equation (5) is > 1, there would be an enhanced temperature dependence of Q which would account for the observed large m values for high T_c materials. For the situation where below $1-Q < 0.05$ the interconnections join very weakly, it can be seen that in the absence of magnetic field the connections with $d_n < 1.34\xi_n$ will contribute little to pinning. As the magnetic field is applied d_n decreases and the number of effective pinning centres would therefore increase. This would explain the nearly constant (or even a slight increase) value of J_c in increasing the magnetic field which would not result in the conventional pinning. The intragrain $J_c(B)$ data on the powdered samples of 123 compound, showing J_c slightly increasing with B, do seem to suggest that the proximity pins are present even in individual grains, probably in the form of twin boundaries or regions of composition fluctuations.

Apart from the restrictions on J_c arising from weak links, proximity connections and anisotropies the high temperature superconductors seem to possess some inherent difficulties for realizing optimum pinning. Owing to their unusually large values of the thermodynamic critical field $B_c(0)$ the FLL is extremely rigid. The shear modulus C_{66} of the FLL is given by Labusch[38],

$$C_{66} = \frac{B_c^2k^2(2k^2-1)}{4\pi[1+1.16(2k^2-1)]^2} \times 0.48\left(1-\frac{B}{B_{c_2}}\right)^2. \quad (6)$$

Since $C_{66} \propto B_c^2$, the FLL in the 123 compound is expected to be at least by an order of magnitude more rigid than in Nb_3Sn. A rigid FLL is difficult to pin as the flux lines are not elastically compliable to adjust themselves to take the energetically most favoured positions at the pinning centres. An exceedingly stiff FLL necessitates the presence of particularly stronger pinning centres which are presumably not there in the ceramic superconductors. When pinning is weak the flux creep becomes a relevant factor in suppressing J_c, giving

$$(F_\perp)_c = [E_p - KT\ \ln(R_o/R_c)](X_p \cdot V_p)^{-1}, \quad (7)$$

where E_p is the height of the pinning energy barrier which is related to F_p through X_p which has the dimension of length, and R_c is the minimum detectable creep rate. When F_p is small and T is large the pins get broken at a lower value of J_c. Studies of the Bitter patterns of the mixed state in 123 compounds have revealed the FLL to become highly unstable as liquid nitrogen temperature is approached[39]. Clearly, more efforts are needed to understand the 'current' problems of high temperature superconductors before they are solved.

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