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On Pinning of Superconducting Flux Lines by Grain Boundaries*

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A simple model is developed which describes the reduction of the mean free path of conduction electrons in metals near a grain boundary. This leads to a decrease of the self-energy of flux lines in a layer which is considerably thicker than the perturbed zone of the boundary itself. The model yields pinning forces which agree, within an order of magnitude, with recent measurements on niobium bicrystals, and with observed values of grain boundary pinning in Nb_3Sn.

1. INTRODUCTION

The critical current in polycrystalline, type II superconductors has frequently been found proportional to the inverse of the mean grain size.[1–11] This is generally interpreted as arising from flux line pinning by grain boundaries. Because of the complicated metallurgical states of the specimens, however, it is usually difficult to distinguish between the acting elementary pinning forces.

Recently Das Gupta *et al.*[12] published measurements of critical currents in niobium bicrystals which, because of the simple geometry, allow a clear separation of grain boundary pinning from surface and bulk pinning. They find elementary pinning forces per unit length of a flux line of about 7×10^{-6} N/m and a symmetric pinning potential on both sides of a plane grain boundary.

None of the various proposed theoretical explanations of grain boundary pinning has achieved general acceptance. Essentially two models have been discussed. The first mechanism, as proposed by Nembach[11] and further

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elaborated by Pande and Suenaga,[13] is based on the stresses emerging from the grain boundary. The second idea, put forward by Campbell and Evetts,[14] takes into account the anisotropy of the self-energy of flux lines. Since the induction in adjacent grains has different crystallographic orientations, this leads to a net pinning force in polycrystals.

Das Gupta *et al.*,[12] however, present a number of convincing arguments against both of these theories. On the one hand, a dislocation wall which forms a symmetric tilt boundary does not give rise to any long-range stress fields which could yield observable pinning forces. The anisotropy model, on the other hand, explains neither the value of the observed pinning force nor the symmetry of the pinning potential. Considering the drastic reduction of the H_{c2} anisotropy with increasing impurity content,[15] it is even more difficult to understand how anisotropy effects could be responsible for grain boundary pinning in compound superconductors.

The present paper shows that the reduction of the electron mean free path due to a grain boundary is effective not only within the perturbed zone of the boundary itself, but in a much thicker layer. The associated variation of superconducting parameters leads to a considerable pinning interaction between grain boundaries and vortices. This idea was first mentioned, although not elaborated quantitatively, by Van Gurp.[2] Our model gives the correct order of magnitude of the pinning force and describes its dependence on induction, temperature, and impurity concentration.

2. MEAN FREE PATH NEAR A GRAIN BOUNDARY

The model does not make any specific assumptions about the microscopic structure of a grain boundary. The only property used is the fact that at a grain boundary the periodicity of the lattice is disturbed drastically so that conduction electrons cannot pass it without being scattered. We only consider the simplest case of diffuse isotropic scattering.

For the quantitative treatment, a plane grain boundary in the y–z plane is considered. In order to estimate the mean free electron path at a distance x from the grain boundary we consider an electron passing through a certain point A at that distance. An effective mean free path is defined as the mean distance l which this electron will travel before suffering its next collision (which is the same as the mean distance which it has traveled since its preceding collision).

Let us first calculate the mean free path l_θ along one arbitrary straight line which passes through A and crosses the grain boundary forming an angle $\pi/2-\theta$ with its plane. Calling l_b the mean free path in the bulk, the probability for the electron to travel a distance r without being scattered is $\exp(-r/l_b)$ if the grain boundary is not within the distance r; otherwise this

probability is zero. Using the abbreviation

$$s = x/\cos\theta \qquad \text{for} \quad \theta < \pi/2 \tag{1}$$

where s is the distance between A and the grain boundary along the line considered, the distribution function takes the form

$$P(r) = \begin{cases} P_1(r) = (1/l_b)\int_0^r \exp(-r'/l_b)\,dr' & \text{for} \quad 0 < r < s \\ P_2(r) = 1 - P_1(s) & \text{for} \quad r = s \\ P_3(r) \equiv 0 & \text{for} \quad s < r < \infty \end{cases} \tag{2}$$

Thus the mean free path l_θ becomes

$$l_\theta = (1/l_b)\int_0^s r \exp(-r/l_b)\,dr + s[1 - P_1(s)] \tag{3}$$

which after a straightforward calculation yields

$$l_\theta = l_b[1 - \exp(-s/l_b)] \tag{4}$$

Half of the electrons will travel into directions with this reduced mean free path (those with $\theta < \pi/2$); the mean free path along the directions $\theta > \pi/2$ remains unchanged, equal to l_b. Averaging over the solid angle 4π

$$l = \tfrac{1}{2}\int_0^{\pi/2} l_\theta \sin\theta\,d\theta + \tfrac{1}{2} l_b \int_{\pi/2}^{\pi} \sin\theta\,d\theta \tag{5}$$

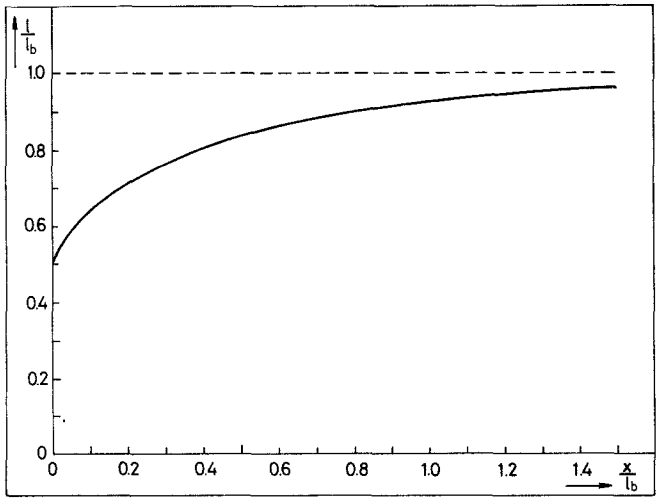

Fig. 1. Variation of the local mean free path l with the distance x from the grain boundary both normalized to the bulk mean free path l_b.

the effective mean free path l of all outgoing electrons becomes finally

$$l = l_b - \tfrac{1}{2} l_b \exp(-x/l_b) - \tfrac{1}{2} x \,\mathrm{Ei}(-x/l_b) \tag{6}$$

$\mathrm{Ei}(u)$ is the exponential integral.[16]

Figure 1 shows l/l_b as a function of x/l_b. Since $\mathrm{Ei}(u)$ diverges logarithmically for $u \to 0$, the last term in Eq. (6) vanishes for $x \to 0$. Thus the mean free path at the boundary becomes half of its bulk value,

$$l \to \tfrac{1}{2} l_b - \tfrac{1}{2} x \, ln \, (x/l_b) \qquad \text{for} \quad x \ll l_b \tag{7}$$

For large negative arguments ($u \ll -1$), on the other hand, the exponential integral can be approximated by $\exp(u)/(-u)$. For large distances x, the mean free path l thus approaches l_b asymptotically as

$$l \to l_b[1 - \exp(-x/l_b)] \qquad \text{for} \quad x \gg l_b \tag{8}$$

3. COHERENCE LENGTH, FLUX LINE ENERGY, AND PINNING

In order to estimate the self-energy of flux lines in a region with varying mean free path Goodman's interpolation formula[17] may be used, which relates the Ginzburg–Landau coherence length ξ with the mean free path l,

$$\xi = \xi_0/(1 + 1.44\xi_0/l)^{1/2} \tag{9}$$

Provided that this relation remains locally valid even for varying mean free path, a combination of (6) and (9) yields a spatially varying coherence length

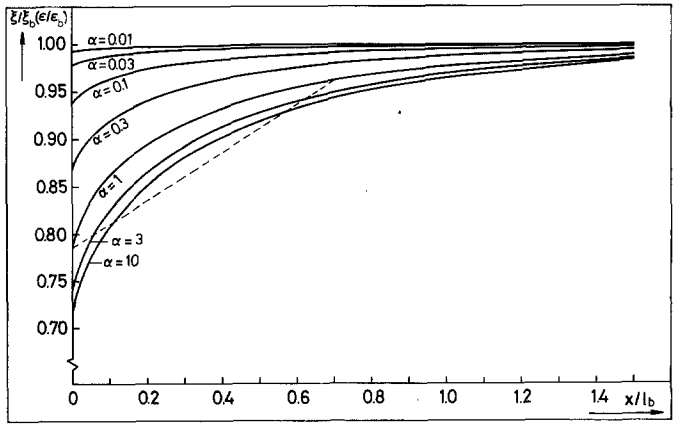

Fig. 2. Variation of the coherence length ξ relative to its bulk value ξ_b with the distance x from the grain boundary, for different impurity parameters α.

ξ, which is shown in Fig. 2 for different impurity parameters $\alpha = 0.882\,\xi_0/l_b$. The reduction of ξ relative to its bulk value ξ_b, is weak for metals of high purity and strong for impure materials. In rather pure niobium, e.g., with a residual resistivity ratio $\Gamma = 1500$ ($\alpha = 10^{-2}$), the reduction of ξ at the boundary is only 0.8% as compared to its bulk value, whereas it amounts to almost 30% in very impure niobium ($\Gamma = 2$, $\alpha = 10$).

For low and medium purities, the main part of the variation takes place for $x \lesssim \xi_b$. Therefore the self-energy of a flux line at a grain boundary is mainly affected through its core. The core energy, defined as the product of the "core cross section" $S\,(=\pi\xi_b^2$ in the bulk) and the condensation energy density, $\mu_0 H_c^2/2$, is accordingly reduced at a parallel grain boundary.

For a flux line parallel to the z direction, it can be estimated that its "radius" in the x direction remains almost unchanged relative to its bulk value ξ_b, whereas in the y direction it is equal to the local value of $\xi(x)$. Therefore an elliptical cross section should be a reasonable approximation to the real flux line core, with an area $S = \pi\xi_b\xi(x)$. Still using strictly local relations, the core energy ε_c relative to its bulk value ε_{cb} would become

$$\varepsilon_c/\varepsilon_{cb} = \xi(x)/\xi_b \tag{10}$$

Thus the curves of Fig. 2 could be interpreted directly as sections through the valleys of the pinning potential along a parallel grain boundary. At this point, however, it is necessary to discuss the validity and the limitations of this local model. Undoubtedly, a strictly local value of the coherence length (and of the mean free path) cannot be used if it varies spatially over short distances. This is best illustrated by the infinite slope of the curves of Fig. 2 for $x \to 0$, which is due to the logarithmic term in Eq. (7). Thus something like an average over the core of a flux line should be used instead.

For a first estimation of the elementary pinning force f_{pL} of a flux line in z direction, the difference in its self-energy between the two positions at $x = 0$ and at $x = \xi_b$, divided by ξ_b, is caluclated. This corresponds to a partial substitution of the curved potentials in Fig. 2 by straight lines (the dashed line in Fig. 2 illustrates this for $\alpha = 1$). In dimensionless form, the elementary pinning force $\tilde{f}_{pL}$ thus becomes

$$\tilde{f}_{pL} = \frac{2f_{pL}}{\pi\xi_0\mu_0 H_c^2} = \frac{\xi_b}{\xi_0}\left\langle\frac{d\xi}{dx}\right\rangle \tag{11}$$

Figure 3 shows this pinning force as a function of the logarithm of the impurity parameter α. The most interesting feature of this curve is the maximum of $\tilde{f}_{pL}$ at $\alpha \cong 1.4$. In very pure metals, $d\xi/dx$ is very small, and the pinning force of grain boundaries also becomes small. With increasing impurity, f_{pL} initially increases together with the mean slope. In very impure

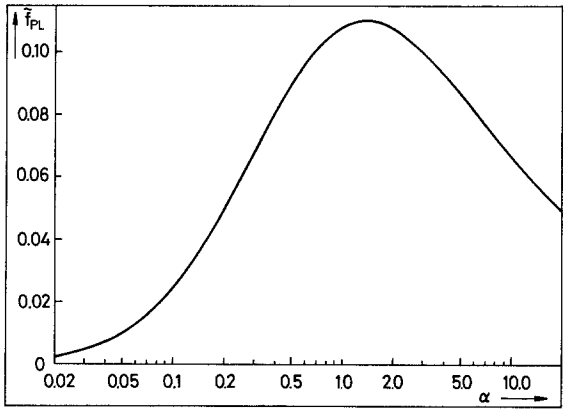

Fig. 3. Reduced elementary pinning force $\tilde{f}_{pL} = 2f_{pL}/\pi\xi_0\mu_0 H_c^2$ as a function of the logarithm of the impurity parameter α.

materials, on the other hand, the coherence length in the bulk ξ_b is already so small that its further reduction due to the boundary has little effect.

4. DISCUSSION

Das Gupta *et al.*[12] observed in niobium a typical elementary pinning force of 7×10^{-6} N/m. The two specimens studied had residual resistivity ratios of 97 and 76, respectively, which correspond to values of impurity parameter α of 0.15 and 0.2. As expected from the model, the critical current of the purer specimen was lower than the critical current of the more impure one. For these values of α, the present model predicts pinning forces of 5.3×10^{-5} and 6.9×10^{-5} N/m [with $\xi_0 = 430$ Å, $H_c = 1.27\times10^5$ A/m = 1.6 kOe (ref. 18)].

In Nb_3Sn, with $\xi_0 = 50$ Å and $H_c = 6.4\times10^5$ A/m (8 kOe), the calculated pinning forces range from 1×10^{-4} to 4.4×10^{-4} N/m for $\alpha > 0.1$. The observed values are between 6×10^{-5} and 1×10^{-4} N/m (α not given).[8–10]

The fact that the observed pinning forces are smaller than those calculated by the present model up to a factor of 10 can be caused by different reasons. First, the model may still suffer from an overstress of locality, despite the averaging procedure described in Section 3. Averaging not only over the slopes but, in any reasonable way, already over the absolute values of l or ξ, would reduce considerably the calculated values of the elementary pinning forces, without affecting the general properties of the curve of Fig. 3. Second, the present model provides a sort of upper limit for the pinning forces in that it assumes that all electrons are scattered at the

grain boundary into random directions. If this scattering probability was less than 1, the pinning effect would be smaller, in the direction of the experimental data. On the other hand, Das Gupta *et al.*[12] had to assume ideal conditions (a single dense row of straight, parallel flux lines interacting with an exactly plane and parallel boundary) in deducing the numerical value of the pinning force from their critical current measurements.

The calculations of Section 3 were made for single flux lines. Thus the results should be correct for low and medium magnetic fields. They have to be modified, however, for higher flux densities when the flux line cores begin to overlap. In a first approximation, this can be done by inclusion of a factor $\langle|\psi/\psi_\infty|^2\rangle$ into the expression for the core energy. This modifies the pinning force by the same factor.

As shown by Abrikosov,[19] the factor $\langle|\psi/\psi_\infty|^2\rangle$ decreases linearly to zero when the mean flux density approaches B_{c2}. Thus it is expected that the elementary pinning force is independent of the applied field for low and medium fields, and decreases linearly to zero near the upper critical field.

Combining the results of Das Gupta *et al.* (Fig. 5 of Ref. 12) with the magnetization data of Finnemore *et al.*,[18] we obtain the points of Fig. 4. The elementary pinning force indeed is approximately constant for inductions $\mu_0 H \leq 0.2$ T, and decreases for higher fields. Unfortunately no data exist for fields very close to H_{c2}, because of perturbations by the peak effect.[12]

The temperature dependence may be estimated by the following considerations: From (11), the pinning force depends on the temperature-dependent quantities H_c, ξ_b, and $\langle d\xi/dx\rangle = (d\xi/dl)\langle dl/dx\rangle$. It follows from (9) that

$$d\xi/dl \propto \begin{cases} \xi_b^2 & \text{for } \xi_0 \ll l_b \quad \text{pure limit} \\ \xi_b & \text{for } \xi \gg l_b \quad \text{dirty limit} \end{cases} \tag{12}$$

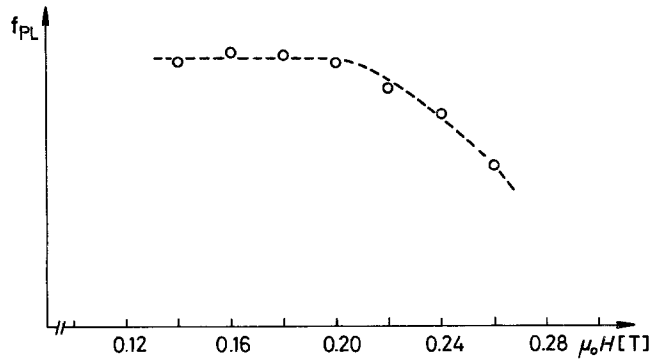

Fig. 4. Observed field dependence of the elementary pinning force in niobium of medium purity (calculated from Refs. 12 and 18).

On the other hand, the upper critical field H_{c2} is proportional to ξ_b^{-2}.[20] Thus it is expected that the scaling law will be of the form

$$f_{pL} = H_c^2(T) H_{c2}^{-n}(T) f(B/B_{c2}) \tag{13}$$

with n between 1.5 (pure limit) and 1.0 (dirty limit).

5. CONCLUSION

Despite the simple assumption applied in the development of the model, the comparison with experimental results shows agreement not only in the order of magnitude of the elementary pinning force between a flux line and a parallel grain boundary but also in its field and impurity dependence. This is an encouraging result for further experimental tests of the model, preferably in bicrystals. Measurements of the impurity dependence of the pinning force seem to be especially interesting.

At the same time one should try to justify the model itself by a microscopic theory of the space-dependent mean free electron path near a grain boundary, as well as by an improved calculation of the self-energy of flux lines in such a region.

It is expected that the described mechanism also contributes, at least partially, to the surface barrier for flux lines, since the mean free electron path near the surface should be reduced in a similar way.[2]

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REFERENCES

1. J. J. Hanak and R. E. Enstrom, in *Proc. of the 10th Conf. on Low Temperature Physics, LT-10, Moscow* (1967), Vol. IIA, p. 10.
2. G. J. Van Gurp, *Phil. Res. Rep.* **22**, 10 (1967).
3. H. Conrad, L. Rice, E. L. Fletcher, and F. Vernon, *Mat. Sci. Eng.* **1**, 360 (1967).
4. A. T. Santhanam, *J. Mat. Sci.* **11**, 1099 (1976).
5. K. J. Gifkins, C. Makeed, and W. A. Rachinger, *Scripta Met.* **2**, 141 (1968).
6. G. R. Love and C. C. Koch, *Appl. Phys. Lett.* **14**, 250 (1969).
7. E. Nembach and K. Tachikawa, *J. Less-Comm. Met.* **19**, 359 (1969).
8. R. M. Scanlan, W. A. Fietz, and E. F. Koch, *J. Appl. Phys.* **46**, 2244 (1975).
9. B. J. Shaw, *J. Appl. Phys.* **47**, 2143 (1976).
10. A. W. West and R. D. Rawlings, *J. Mat. Sci.* **12**, 1862 (1977).
11. E. Nembach, *Z. Metallk.* **61**, 734 (1970).
12. A. Das Gupta, C. C. Koch, D. M. Kroeger, and Y. T. Chou, *Phil. Mag.* B **38**, 367 (1978).
13. C. S. Pande and M. Suenaga, *Appl. Phys. Lett.* **29**, 443 (1976).

14. A. M. Campbell and J. E. Evetts, *Adv. Phys.* **21**, 199 (1972).
15. E. Seidl and H. W. Weber, in *Anisotropy Effects in Superconductors*, H. W. Weber, ed. (Plenum Press, New York, 1977).
16. E. Jahnke, F. Emde, and F. Lösch, *Tables of Higher Functions* (B. G. Teubner, Stuttgart, 1960).
17. B. B. Goodman, *Rep. Prog. Phys.* **29**, 445 (1966).
18. D. K. Finnemore, T. F. Stromberg, and C. A. Swenson, *Phys. Rev.* **149**, 231 (1966).
19. A. A. Abrikosov, *Zh. Eksp. Theor. Fiz.* **32**, 1442 (1957).
20. P. G. de Gennes, *Superconductivity in Metals and Alloys* (Benjamin, New York, 1966).