Pinning forces and defects as measured by critical currents in amorphous superconducting materials

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Analysis of the available data on critical currents in amorphous superconductors shows a striking similarity in the defects responsible for the pinning force in all samples in which two-dimensional flux pinning can be observed. Order-of-magnitude calculations show that this is consistent with the existence of disclinations in glasses as has been proposed in different theoretical papers by Rivier, Nelson, Kleman, and Sadoc. The evolution of pinning forces with annealing also shows a similar behavior in all the published data, and the trend is toward a state with fewer or smaller disclinations. If disclinations are identified with two-level systems (TLS) this trend is consistent with a decrease of the scattering of phonons by TLS which is observed upon annealing in thermal conductivity measurements.

Pinning forces in superconductors are, in principle, a sensitive way of studying the defects present in a given material, but the interpretation of the experimental results is not usually straightforward because of the theoretical complexity of the problem. One needs to know the behavior of the flux-line lattice (FLL), the individual pinning force f_p, and what summation rule to use for the calculation of the average pinning force $\langle f_p \rangle$. Thus full quantitative agreement between calculations and measurements of the resultant pinning force F_p are difficult to achieve. One instance in which good agreement between theory and experiment has been reported is when the conditions for two-dimensional (2D) collective flux pinning have been fulfilled. Measurements on amorphous superconductors[1-3] have been quantitatively explained by means of the theory of collective flux pinning proposed by Larkin and Orchivnikov[4] (LO). Amorphous materials appear to be good candidates for observing collective pinning because it seems that they have a large density of weak pinning centers, but although the agreement between theory and experiment is good, the microscopic nature of the pinning centers is not clear because of the lack of a better understanding of the structure of glassy materials and of the defects present in them.

In the experiments where 2D pinning has been observed the curves of the measured pinning force F_p against the reduced magnetic field $b \equiv H/H_{c2}$ were fitted assuming that the defects responsible for the pinning are dislocation loops. The interaction of the strain field of the FLL with the stress field of the dislocation loops[5-7] produces an elementary pinning force

$$\langle f_p \rangle = \tfrac{1}{2} n_d f_p^2 \propto b(1-b) \ , \tag{1a}$$

or[3]

$$\langle f_p \rangle \propto b(1-b)(1-gb) \ , \tag{1b}$$

where n_d is the number of defects, $\langle f_p \rangle$ an average pinning force, and f_p the pinning force of a single dislocation loop.

Other defects, such as voids, give an elementary pinning force[7,8]

$$\langle f_p \rangle \propto b^3(1-b) \ , \tag{1c}$$

whose field dependence is not in agreement with the experimental data.

Although the formulas for dislocation loops give a good fit for the measurements, it is far from clear how dislocation loops as such can exist in an amorphous material, and no microscopic model for the defects responsible for the pinning has been given so far. If one uses the quasi-dislocation-loop concept and accepts the formulas at face value, some tentative conclusions as to the density and diameter of the loops can be drawn. The measured pinning force is (2D LO theory[4])

$$F_p = W(0)/da_0 C_{66} \ , \tag{2}$$

where d is the sample thickness, a_0 is the lattice constant of the FLL, and C_{66} is the shear modulus of the FLL given by Brandt[9] in the large-K limit:

$$C_{66} = H_c^2/4\pi \left[\frac{K(T_c)}{K(T)} \right]^2 \frac{b(1-b)^2}{4} (1-0.29b) \ , \tag{3}$$

where H_c is the thermodynamic critical field and $K(T)$ is the temperature-dependent Ginzburg-Landau parameter.

$$W(0) = \langle f_p \rangle \cong \tfrac{1}{2} n_d f_p^2 \tag{4}$$

and the elementary pinning force can be written as[10]

$$f_p = 0.43(2\pi/a_0)\mu b^* \frac{(1+\nu)}{(1-\nu)} d\varepsilon_V (1-b)\pi D^2/4 \ , \tag{5}$$

where μ is the shear modulus of the material, b^* is the Burgers vector of the dislocation, ν is Poisson's ratio, D the diameter of the loop, and $d\varepsilon_V$ is the volume dilation which can be related to the pressure dependence of the critical field by[1]

$$d\varepsilon_V = (H_c/8\pi) dH_c/dP \ . \tag{6}$$

The FLL spacing is related to the applied field H by

$$a_0=(2/\sqrt{3})(\phi_0/H)^{1/2}=(2/\sqrt{3})\phi_0^{1/2}/b^{1/2}H_{c2}^{1/2} , \quad (7)$$

where $\phi_0=2.07\times10^{-7}$ G cm^2 is the flux quantum.

From the above equations one obtains

$$n_dD^4=\frac{256\phi_0^{3/2}}{3\sqrt{3}\pi^3}\frac{[K(T_c)/K(T)]^2}{\{0.43\mu b^*[(1+\nu)/(1-\nu)]dH_c/dP\}^2}\times\frac{F_p(b)}{H_{c2}^{3/2}}d\frac{(1-0.29b)}{b^{1/2}} , \quad (8)$$

taking $\mu=3\times10^{11}$ dyn cm^{-2}, $b^*=2.8\times10^{-8}$ cm, and $\nu=0.39$ as was assumed by Kes and Tsuei,[1] and using $dH_c/dP=6.2\times10^{-9}$ G cm^{2}dyn^{-1}, which is the value obtained by Olsen and Rohrer[11] for lanthanum:

$$n_dD^4=5.75\times10^{-2}F_p/H_{c2}^{3/2}d(1-0.29b)/b^{1/2} , \quad (9)$$

with n_dD^4 in cm if cgs units are used for d, F_p, and H_{c2}.

In Table I the values of n_dD^4 are tabulated for all the materials for which the 2D theory is valid and a "universal" behavior is seen, that is, all materials have remarkably close values for the product n_dD^4.

Although this is only a small selection of amorphous superconductors, the materials are different with a range of T_c's that goes from 7.69 to 2.8 K, and they are prepared by different techniques: the Zr-based alloys are melt spun and the other samples are sputtered. The similarity in behavior could be explained if the defects were not just a result of the way in which the samples were prepared but rather a fundamental property of the glass itself.

Recent theoretical results of Rivier,[13,14] Rivier and Duffy,[15] Kleman and Sadoc,[16] and Nelson[17] have focused attention on disclinations as important defects of amorphous solids which are absent in normal crystalline materials because their elastic energy is prohibitively high. The models proposed do not agree in detail, but from the point of view of the pinning forces it is interesting to test the concept of disclinations against the experimental results. Disclinations would replace dislocations as a source of stress and they could interact with the FLL to produce a pinning force.

In the model of Rivier[13-15] disclinations are identified with odd lines, that is, continuous lines threading the odd-numbered faces of a Voronoi partition of the solid which either form loops or end in the boundaries of the sample. It can be shown[13] that odd-numbered faces are not present in isolation. In Rivier's model it is assumed that the odd faces are not too abundant and that the distribution of odd lines is semidilute. Nelson,[17] on the other hand, assumes that in glasses there is a tendency toward icosahedral order and thus five-sided faces are the norm in the Voronoi partition. Odd lines still form loops but they are very small, and it is the faces with an even number of sides which are connected by disclination lines. Thus, in both cases the geometry of the defects is indeed that of loops or lines and one can compare the strains produced by them with that of dislocation loops to see if they could be responsible for the pinning force.

Rivier[14] calculates for the elastic energy of a semidilute distribution of loops as follows:

$$E_{OL}=F[-An_d\ln(n_d)+Bn_d] , \quad (10)$$

where F is the number of faces, n_d is the density of odd lines, B is a core energy, and $A=kT_g$ with T_g the glass temperature. The interaction potential of the disclination loops assumes the form $1/|x-x'|$ which is of the same form as the interaction potential between dislocations.

On the other hand, the strain energy of a dislocation loop is approximately[18]

$$e_D=b^{*2}G\ln(R/r_0)\pi D+e_{\text{core}} , \quad (11)$$

where G is an elastic constant, usually of the order of 10^{11}–10^{12} dyn cm^{-2}, b^* is the Burgers vector of the dislocation, D is the diameter of the loop, R is a distance of the order of the sample dimensions, and r_0 is of the order of a few atomic distances.

If it is assumed that the core of a disclination and a dislocation have a similar energy and that the Burgers vector is of the order of an interatomic distance which is of about the same order of magnitude as the deformation at the core of the disclination, one can equate from Eqs. (10) and (11) that

$$E_{OL}=n_de_D$$

to obtain an order of magnitude estimate for D. It is necessary to assume a value for n_d to obtain D and one can use a further important result of Ref. 13, the identification of odd lines with the two-level systems (TLS) responsible

TABLE I. Data of pinning forces for samples which show 2D collective flux pinning. The product of the number of "quasidislocation loops" and their diameter raised to the fourth power is calculated from Eq. (9) in the text. The values of D are estimated assuming a density of disclinations equal to that of TLS, i.e., one every 10^5 atoms.

Material	Preparation technique	T_c (K)	t	b	$F_p(b)$ (10^3 dyn cm^{-3})	H_{c2} (10^3 G)	d (10^{-4} cm)	n_dD^4 (10^{-8} cm)	n_d (10^{17} cm^{-3})	D (Å)	Refs.
Nb_3Ge[a]	Sputtering	4.00	0.7	0.4	16.1	23.6	0.46	1.64	5.19	42	1
Nb_3Ge[b]	Sputtering	3.99	0.7	0.4	6.49	24.0	1.24	1.74	5.19	43	1
Nb_3Si	Sputtering	3.20	0.65	0.4	8.10	16.6	0.22	0.67	5.40	33	1
Mo_3Si	Sputtering	7.69	0.75	0.5	57.20	35.7	0.46	2.70	6.28	46	1
$Zr_{70}Cu_{30}$	Melt spinning	2.80	0.65	0.9	0.85	22.6	12.00	1.34	5.25	40	2,12
Zr_3Rh	Melt spinning	4.24	0.67	0.9	2.0	37.4	12.00	1.48	5.10	41	3

[a]Corresponds to sample 191B, Ref. 1.

[b]Corresponds to sample 175A, Ref. 1.

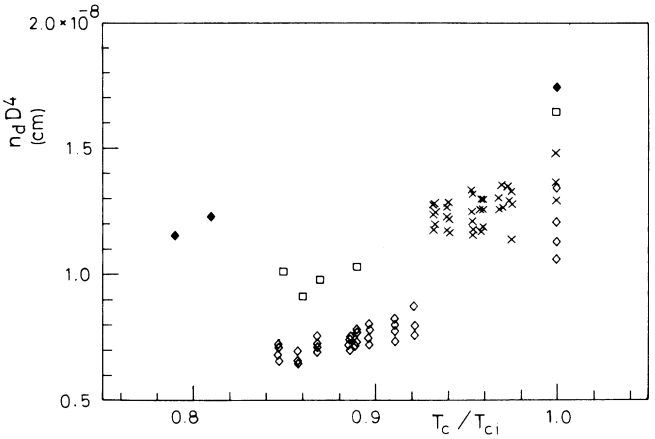

FIG. 1. Evolution of defects with annealing as a function of critical temperature divided by the T_c of the "as-quenched" samples. Crosses, Zr_3Rh (Ref. 3); open diamonds, $Zr_{75}Cu_{25}$ (Refs. 2 and 18); squares, Nb_3Ge (sample 191B, Ref. 1); filled diamonds, Nb_3Ge (sample 175A, Ref. 1). The data of Zr_3Rh and $Zr_{75}Cu_{25}$ correspond to different measuring temperatures.

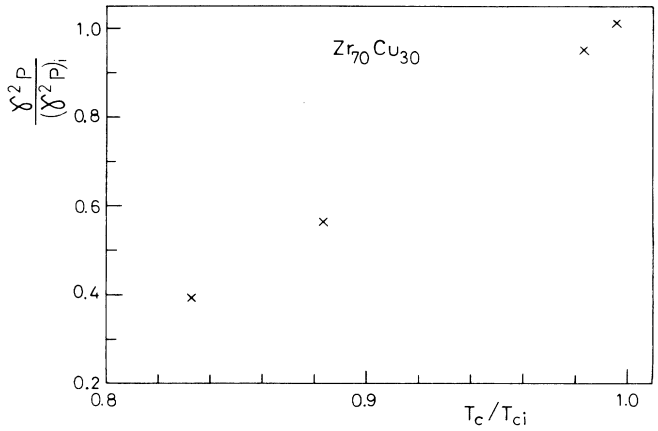

FIG. 2. Evolution of the product of density of states times phonon-TLS coupling constant as a function of temperature. Both quantities are normalized by the value of the "as-quenched" samples. Data are taken from Refs. 27 and 28.

for the characteristic behavior of the specific heat, thermal conductivity, sound attenuation, etc. of glasses at low temperatures. The density of TLS is of the order of one every 10^5 or 10^6 atoms,[19] and this is the density assumed for the disclination loops.

Taking $G=10^{11}$ dyn cm^{-2}, $b^*=10^{-8}$ cm, $R/r_0=10^5$, $n_d=10^{-5}$, $T_g=800$ K, and $F=\frac{1}{2}\langle f\rangle C$, where $\langle f\rangle=14$ is the average number of faces and C the number of cells (equal to the number of atoms), one obtains

$$D=30\times10^{-8}\text{ cm}=30\text{ Å}\ . \tag{12}$$

If one uses $n_d=10^{-5}$ per atom in Table I, an estimate of the diameter of the "quasidislocation loops" can be obtained as can be seen in column 9 of Table I where the values of D for all samples are around 40 Å. Therefore the pinning forces measured are compatible with a picture of the glass with disclinations as the main defects, at least within the very coarse approximations used here.

Disclination lines are also topologically stable and this should be reflected in the behavior of the pinning force with annealing. In Fig. 1 the evolution of n_dD^4 with annealing is plotted as a function of T_c/T_{ci}, where T_c is the critical temperature and T_{ci} is the T_c of the "as quenched" samples. It can be seen that the evolution of the defects is correlated with the critical temperature and again a similar behavior is followed by all the samples. The change in the defects is less than the change in the measured pinning force because the pinning forces reflect not only the changes in the pinning centers but also changes in the superconducting matrix (such as T_c and H_{c2}) and possibly changes in the pinning regime, which can change from two dimensional to three dimensional.[20] It is possible too that other types of defects can also contribute and thus raise the value of F_p. This is clearly the case when crystalline precipitates are present, as in the measurements of Clemens, Johnson, and Bennet[21] on $(Mo_{0.6}Ru_{0.4})Si_{10}B_{10}$ where an increase in the pinning force due to the precipitates is seen. The samples used, however, are 50 μm thick so they are not in the 2D regime and the analysis of their defects is not so straightforward. Other measurements of critical currents published in the literature[22–26] show results clearly outside the 2D regime. Most of the samples are more than 30 μm thick and they do not fulfill the 2D criterion nor possibly the requirements for collective flux pinning.

Results of thermal conductivity with annealing are available for $Zr_{70}Cu_{30}$ and an increase of the thermal conductivity is observed with heat treatment.[27,28] The increase is due to a decrease of the scattering of phonons by the TLS, which is proportional to γ^2P where γ^2 is the coupling constant of the phonons and TLS and P is the density of states of the TLS at low energies. In a plot of $\gamma^2P/(\gamma^2P)_i$ vs T_c/T_{ci} (Fig. 2) a decrease which is roughly linear can be seen. This is consistent with an identification of TLS and odd lines although the relative decrease in γ^2P is about twice that of n_dD^4 for $Zr_{70}Cu_{30}$. The discrepancy could be absorbed by a change in γ^2, but too detailed a comparison makes little sense within the present state of experimental and theoretical knowledge.

In conclusion, the present analysis shows that the pinning characteristics in a variety of amorphous alloys are remarkably similar. This is consistent with a view which identifies the defects as essential constituents of glasses, either arising from the impossibility of filling ordinary space with tetrahedra,[17] as a projection from an ideal space where such a filling is possible,[16,29] or as a result of the constraints imposed by space filling requirements and the maximization of entropy.[14] A more detailed theoretical description could perhaps give a unified picture of the structure of defects in amorphous materials, but within the rather simplified analysis given here the existence of disclinations in glasses seems to account for the experimental data in a semiquantitative fashion.

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[1]P. H. Kes and C. C. Tsuei, Phys. Rev. B **28**, 5126 (1983).
[2]E. J. Osquiguil, V. L. P. Frank, and F. de la Cruz, Solid State Commun. **55**, 227 (1985).
[3]J. Luzuriaga, C. D'Ovidio, and F. de la Cruz, Solid State Commun. **57**, 753 (1986).
[4]A. I. Larkin and Yu. Orchivnikov, J. Low Temp. Phys. **34**, 409 (1979).
[5]C. S. Pande, Appl. Phys. Lett. **28**, 462 (1976).
[6]G. Kluge and V. Rudat, Zh. Eksp. Teor. Fiz. **62**, 1192 (1974) [Sov. Phys. JETP **38**, 783 (1974)].
[7]E. J. Kramer, J. Appl. Phys. **49**, 742 (1978); Philos. Mag. **33**, 331 (1976).
[8]A. M. Campbell and J. E. Evetts, Adv. Phys. **21**, 199 (1972).
[9]E. H. Brandt, Phys. Stat. Solidi (b) **77**, 551 (1976).
[10]R. Labusch, Phys. Rev. **170**, 470 (1968).
[11]J. L. Olsen and H. Rohrer, Helv. Phys. Acta **33**, 872 (1960).
[12]V. L. P. Frank, Ph.D. thesis, Universidad Nacional de Cuyo (unpublished).
[13]N. Rivier, Philos. Mag. A **40**, 859 (1979).
[14]N. Rivier, in *Amorphous Materials: Modeling of Structure and Properties,* edited by V. Vitek (The Metallurgical Society, Warrendale, PA, 1982).
[15]N. Rivier and D. M. Duffy, J. Phys. (Paris) **43**, 293 (1982).
[16]M. Kleman and J. F. Sadoc, J. Phys. (Paris) Lett. **40**, L569 (1979).
[17]D. R. Nelson, Phys. Rev. B **28**, 5515 (1983).
[18]N. W. Ashcroft and N. D. Mermin, *Solid State Physics* (Holt, Reinhart and Winston, New York, 1976).
[19]J. L. Black, in *Glassy Metals I,* edited by H. J. Guntherodt and H. Beck (Springer-Verlag, New York, 1981).
[20]E. N. Martinez, E. J. Osquiguil, V. L. P. Frank, and F. de la Cruz, Solid State Commun. (to be published).
[21]B. M. Clemens, W. L. Johnson, and J. Bennet, J. Appl. Phys. **51**, 1116 (1980).
[22]B. M. Clemens and W. L. Johnson, J. Appl. Phys. **53**, 7612 (1982).
[23]N. Toyota, T. Fukase, A. Inoue, Y. Takashashi, and T. Masumoto, Physica B **107**, 465 (1981).
[24]M. Tenhover and B. M. Clemens, J. Phys. D **16**, 1761 (1983).
[25]C. C. Koch, J. O. Scarbrough, D. M. Kroeger, and A. Das Gupta, Appl. Phys. Lett. **37**, 451 (1980).
[26]P. Esquinazi, B. Gillet, R. Steinman, and H. Dussel, J. Non-Cryst. Solids **69**, 17 (1985).
[27]P. Esquinazi, M. E. de la Cruz, A. Ridner, and F. de la Cruz, Solid State Commun. **44**, 941 (1982).
[28]P. Esquinazi, Ph.D. thesis, Universidad Nacional de Cuyo (unpublished).
[29]N. Rivier and A. Lawrence, J. Phys. C **8**, 409 (1985).