Intergrain flux creep in high- T_c superconductors

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Equations describing the intergrain flux creep of ceramic superconductors are derived. They are based on the behavior of small superconducting loops closed by overdamped Josephson junctions. The elementary loops are formed around nonsuperconducting regions between the grains, the loops acting as pinning centers. Despite different pinning mechanisms the equations for the relaxation of the intergrain current density in a static and a slowly varying external magnetic field are formally similar to those of the classical flux-creep theory based on volume pinning.

Soon after the discovery of ceramic superconductors it was proposed that a polycrystalline sample can be described as a positionally disordered system of grains coupled via Josephson weak links.¹ When the sample is exposed to a magnetic field, two kinds of current system will be induced in the superconductor comprising interand intragrain screening currents.² While the electromagnetic behavior of the intragrain system is now quite well modeled, the understanding of the electrodynamics of the intergrain system is far from complete.

Supposing that the grains are in the Meissner state the polycrystalline sample can be modeled as a network of Josephson junctions (JJ), where the current flows in a thin surface layer (of thickness λ) of the grains and crosses the weak links. In the first studies of the behavior of such a network, screening effects due to the magnetic field of the Josephson currents were often neglected.³⁻⁵ Such a simplification can be allowed when studying the behavior of samples comprising a very low critical transport current density, but not for samples with moderate J_c (in excess of 100 A/cm²). Recently there have been published numerical studies of the JJ network magnetization accounting for the magnetic energy of the screening currents.⁶⁻⁹ The results of these studies are in good agreement with the experimental results of the flux distribution in polycrystalline ceramic superconductors published by the authors.^{10,11}

In this paper we present a model describing the intergranular flux creep of ceramic superconductors. The model is based on the concept of a network of small superconducting current loops, each containing resistively shunted Josephson junctions and having a finite loop inductance. The elementary JJ loops are formed around nonsuperconducting regions between the grains, the loops acting as pinning centers. Thus, screening effects and irreversibilities play a significant role in the behavior of the system. The relaxation of induced screening currents in the elementary loops is considered both in a static applied field and during the sweep of an applied field.

Let us first consider a superconducting loop of inductance L closed by an ideal Josephson junction shunted by a small capacitance C and a resistance R. The size of the junction is supposed to be so small that the effects of the self-field on the junction current can be neglected. The flux ϕ threading the ring is related to the applied flux ϕ_x by

$$\phi = \phi_x - Li_c \sin(2\pi\phi/\phi_0) - LC\ddot{\phi} - L\dot{\phi}/R , \qquad (1)$$

where i_c is the maximum current of the junction and ϕ_0 is the flux quantum.¹² Equation (1) describes the flux change of a JJ loop. Rearranging the terms and dividing by L results in the current equation

$$(\phi - \phi_x)/L + i_c \sin(2\pi\phi/\phi_0) = -C\ddot{\phi} - \dot{\phi}/R , \qquad (2)$$

where the left-hand side represents the induced screening current and the tunneling current, respectively. These currents may be considered the result of an energy potential function $U(\phi, \phi_x)$:

$$U(\phi, \phi_x) = (\phi - \phi_x)^2 / 2L + E_0 [1 - \cos(2\pi\phi/\phi_0)], \qquad (3)$$

where the first term on the right-hand side describes the magnetic energy created by the loop and the second term, with $E_0 = i_c \phi_0/2\pi$, the Josephson energy of the junction. In Eq. (2) the resistive current $\dot{\phi}/R$ has a damping effect on the flux dynamics, accordingly one can define a damping coefficient $\eta = 1/RC$. The study is restricted to junctions with large η (overdamped junctions), i.e., junctions with almost negligible capacitance. This assumption is reasonable for junctions with a cross-sectional area in the μm^2 range.

The behavior of the system can be better understood by studying its trajectory in $U(\phi, \phi_x)$. Figure 1 shows the potential in normalized units for the case $Li_c = 5\phi_0$. Let us first consider the system in a state, wherein a certain flux amount is trapped in the superconducting loop after the external field has been decreased to zero. The system is locked in the fluxoid potential well, say point A in Fig. 1. The state is constrained by a potential barrier $\Delta U(\phi_x = 0) = U(\phi_B) - U(\phi_A)$. In the absence of energy fluctuations it will stay in this state. However, when exposed to thermal fluctuations of the average level k_BT $(k_BT < \Delta U)$, the system will oscillate with a frequency ω around the minimum of the potential well and ultimately turn to a lower metastable state. If the damping



FIG. 1. Potential surface $U(\phi, \phi_x)$ for a superconducting loop closed by a Josephson junction in the case $Li_c = 5\phi_0$. For the legend see the text.

coefficient is large enough, the system will be retrapped in the next lowest quantum state, point C. In other words, this means the escape of a flux quantum from the loop through a 2π phase-slip process of the junction. In the case of high damping, the lifetime τ associated with the flux remaining in a specific quantum state is^{13,14}

$$\frac{1}{\tau} = \frac{\omega^2}{2\pi\eta} e^{-\Delta U/k_B T} . \tag{4}$$

In the following the amplitude factor, although the frequency ω is slightly different in adjacent quantum states, will be replaced by a constant $\omega_0 = \omega^2/2\pi\eta$, which occasionally is of the order R/L. In this sense the situation is analogous to the classical flux-creep theory,¹⁵ where a constant attempt frequency is used in describing the oscillations of pinned vortices. Because the current and the flux of the loop are coupled together by the inductance, it is convenient to write the barrier height ΔU between adjacent states as a function of the current *i* flowing through the junction:¹⁶

$$\Delta U = E_0 \left\{ \left[1 - \left[\frac{i}{i_c} \right]^2 \right]^{1/2} - \frac{i}{i_c} \cos^{-1} \left[\frac{i}{i_c} \right] \right\}.$$
 (5)

Next, let us consider the magnetization of a zero-fieldcooled system during a slow sweep of the applied field. Assuming that $\dot{\phi}_x \ll \phi_0 \omega_0$ we start from point D, Fig. 1. As ϕ_x is increased the potential barrier decreases and vanishes at point E causing a system transition to point F. Let this critical flux value be ϕ_{xc} . While increased the external field induces a current, which perfectly shields the interior of the loop until $\phi_x = \phi_{xc}$. Then a flux quantum irreversibly penetrates the loop. However, at nonzero temperature a thermally activated transition will occur before ϕ_{xc} .^{13,17} As ϕ_x is slowly increased the potential energy of the system increases along the valley D-E. A transition to F' occurs already at point E' $(\phi_{x(E')} < \phi_{xc})$, where the barrier ΔU is so small that a thermally activated transition is likely to occur. According to Kurkijärvi¹³ the probability of a jump at a lower ϕ_x value increases as the sweep rate decreases, which is an obvious consequence of Eq. (4).

Among the superconducting grains there exist nonsuperconducting regions consisting of nonstoichiometric material and voids. During the magnetization process the screening currents around such regions flow from one grain to another thus forming a superconducting loop with a few Josephson junctions. Provided that $Li_c > \phi_0$ and that the sizes of the junctions are small enough,¹² the situation returns to the case treated above. A recent numerical analysis by De Luca, Pace, and Saggese¹⁸ shows that, in particular, small elementary loops play a key role in the magnetization process of the whole intergranular system. Especially nonidentical parameters will promote the formation of small loops. It has been proposed that the intergranular nonsuperconducting regions act as pinning centers for intergrain currents.^{19,20} Our model of the intergrain flux creep is based on this assumption. The model concentrates on the behavior of a single JJ loop. In reality an intergranular loop is formed from 3-4 grains. However, for simplicity an elementary loop with only one junction is considered. In the theoretical analysis the error is not crucial, flux creep occurs only in one direction, the negative flux density gradient, hence only one JJ at a time works as a drain. Further, it is assumed that the critical state concept for the intergranular system^{7,8,19} is valid, meaning the existence of a flux density gradient extending over several loops. According to common practice in single-loop analysis the local field influence on the maximum Josephson current is neglected.

Let us consider a magnetized polycrystalline sample in a constant field. The intergrain current I in an elementary JJ loop decreases during each period of time τ on the average by

$$\Delta |I| = -\Delta \phi / L \quad . \tag{6}$$

In overdamped junctions only one flux quantum leaves the loop during each phase slip event, i.e., $\Delta \phi = \phi_0$. Based on Eqs. (4) and (6) the rate of the current density change is obtained from

$$\frac{dJ}{dt} = -\frac{\phi_o \omega_0}{AL} e^{-\Delta U(J)/k_B T}, \qquad (7)$$

where J has been substituted from J=I/A, A is the junction area. In order to solve Eq. (7) the shape of the potential barrier $\Delta U(J)$ is simplified by linearizing $\Delta U(J)$ according to

$$\Delta U(J) = E_0 \left[1 - \frac{J}{J_c} \right] \,. \tag{8}$$

An analogous situation exists in type-II superconductors, where the current dependence of the pinning potential often is linearized in a similar way.²¹ Now by combining Eqs. (7) and (8) the current density can be solved

$$J(t) = J(0) \left[1 - \frac{k_B T}{E_0} \ln \left[1 + \frac{t}{\tau_0} \right] \right], \qquad (9)$$

where

$$\tau_0 = \frac{LAk_B TJ_c}{\phi_0 \omega_0 E_0} \tag{10}$$

and J(0) is the local current density at t=0. J(0) is equal to the J_c of Eq. (8). Equation (9) expresses the relaxation of the local intergrain current due to flux creep. Formally it is similar to that of intragrain currents. According to an experimental study of fluxoid quantization of a polycrystalline YBa₂Cu₃O_{7-x} loop, recently published by Oh, Koch, and Gallagher,²² the relaxation of screening currents exhibits a logarithmic time dependence. Sensitive measurements showed that this was due to a logarithmic slowing down of the rate of individual flux quanta hopping through the loop. This can be interpreted as a support for the present intergrain flux-creep model.

The escape of a flux quantum from the loop increases the phase difference of the superconducting order parameter φ between adjacent grain boundaries by 2π . Because the escape occurs in periods of time τ [Eq. (4)], we can estimate the average change rate of φ from¹⁶

$$\left\langle \frac{d\varphi}{dt} \right\rangle \approx \frac{\Delta\varphi}{\Delta t} = \frac{2\pi}{\tau} \ .$$
 (11)

The voltage across the junction is

$$V = \frac{\hbar}{2e} \left\langle \frac{d\varphi}{dt} \right\rangle \approx \frac{\hbar}{2e} \frac{2\pi}{\tau} . \tag{12}$$

The average electric field is obtained by dividing Eq. (12) by the average grain diameter D and accounting for Eq. (4):

$$E(J) = \frac{\phi_0 \omega_0}{D} e^{-\Delta U(J)/k_B T}.$$
(13)

E as a function of *J* is illustrated in Fig. 2. The potential barrier ΔU is of the type given by Eq. (5). The derived expression for the E(J) relation due to intergrain current relaxation is in a good agreement with the experimentally observed one²³ for polycrystalline (Bi,Pb)₂Sr₂Ca₂Cu₃O_x at 77 K. However, those measurements covered only an electric field interval of 3 orders of magnitude.

Next we consider the applied flux to be increased at a constant sweep rate, $\dot{\phi}_x \ll \phi_0 \omega_0$. During small change intervals of ϕ_x the loop flux ϕ remains, in principle, constant. However, thermally activated flux transitions have a decreasing influence on the screening current in the loop. Hereby the actual average change rate of the local current is achieved from

$$\frac{dI}{dt} = \frac{\dot{\phi}_x}{L} - \frac{\phi_0 \omega_0}{L} e^{-\Delta U/k_B T} .$$
(14)

However, the continuous increase of ϕ_x will cause a stepwise change of ϕ . In the long run the average flux ramp $\dot{\phi}$ will be close to $\dot{\phi}_x$. As $dI/dt = (\dot{\phi} - \dot{\phi}_x)/L$ by definition, it follows that $dI/dt \rightarrow 0$. We can now approximate the case by neglecting the left-hand side of Eq. (14) and write

$$\dot{\phi}_x = \phi_0 \omega_0 e^{-\Delta U(J)/k_B T} . \tag{15}$$

We simplify the potential barrier by using Eq. (8). Then by solving Eq. (15) we get an expression for the local current density during a slow sweep of the applied field:



FIG. 2. E(J) characteristic for the intergrain flux creep in the case where $E_0/k_BT=16$, $\omega_0=10^5$ s⁻¹, and the grain size $D=10 \ \mu m$.

$$J = J_c \left[1 + \frac{k_B T}{E_0} \ln \left[\frac{\dot{\phi}_x}{\phi_0 \omega_0} \right] \right] .$$
 (16)

The dependence between current density and the sweep rate $\dot{\phi}_x$ is similar to the one derived by Schnack *et al.*²⁴ for the intragrain current system comprising volume pinning.

It is rather surprising, that the equations for the local intergrain current densities, Eq. (9) for constant ϕ_x and (16) for slowly swept ϕ_x , are formally identical to those of local intragrain current densities. This similarity is one reason, why intergrain systems have been compared to the old concepts of classical type-II superconductors. However, the electrodynamic behavior of type-II superconductors is based on the interaction between Lorentz forces and vortex pinning forces. In the intergrain system of ceramic superconductors comprising small Josephson junctions magnetic flux is present mainly in nonsuperconducting regions inside elementary superconducting loops. The Lorentz-force interaction is highly limited, accordingly the old concepts are not valid. The loops act as pinning centers and the flux creep is due to thermal oscillations in the weak links. Although fluxcreep equations (9) and (16) represent local current densities, they can easily be extended to the whole sample size. Detailed studies of the flux creep in a JJ network are in progress.

In summary, we have presented a model describing thermally activated intergrain flux creep in ceramic superconductors. The model is based on the behavior of superconducting grain loops closed by overdamped Josephson junctions. The equations for the relaxation of the intergrain current density in a static and a slowly varying external flux have been derived. Despite a different pinning mechanism the equations are formally similar to those of the classical flux-creep theory based on volume pinning. This suggests a reevaluation of the current transport concepts in polycrystalline superconductors like high- T_c tapes.

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- ¹K. A. Müller, M. Takashige, and J. G. Bednorz, Phys. Rev. Lett. 58, 1143 (1987).
- ²J. R. Clem and V. G. Kogan, Jpn. J. Appl. Phys. 26, 1159 (1987).
- ³D. Stroud and C. Ebner, Physica C 153-155, 63 (1988).
- ⁴M. Tinkham and C. J. Lobb, in *Solid State Physics*, edited by H. Ehrenreich and D. Turnbull (Academic, San Diego, 1989), Vol. 42.
- ⁵J. R. Clem, Physica C 153-155, 50 (1988).
- ⁶R. De Luca, S. Pace, and B. Savo, Phys. Lett. A **154**, 185 (1991).
- ⁷A. Majhofer, T. Wolf, and W. Dieterich, Phys. Rev. B **44**, 9634 (1991).
- ⁸R. De Luca, S. Pace, and A. Saggese, Cryogenics **32**, 995 (1992).
- ⁹R. De Luca, S. Pace, and G. Raiconi, Phys. Lett. A 172, 391 (1993).
- ¹⁰J. Paasi, M. Polák, M. Lahtinen, V. Plecháček, and L. Söderlund, Cryogenics **32**, 1076 (1992).
- ¹¹J. Paasi, M. Polák, M. Lahtinen, J. T. Eriksson, and V. Plecháček, IEEE Trans. Appl. Supercond. 3, 1378 (1993).
- ¹²A. Barone and G. Paterno, *Physics and Applications of the Josephson Effect* (Wiley, New York, 1982), Chap. 12.

- ¹³J. Kurkijrvi, Phys. Rev. B 6, 832 (1972).
- ¹⁴T. A. Fulton and L. N. Dunkleberger, Phys. Rev. B 9, 4760 (1974).
- ¹⁵P. W. Anderson and Y. B. Kim, Rev. Mod. Phys. 36, 39 (1964).
- ¹⁶P. A. Lee, J. Appl. Phys. 42, 325 (1971).
- ¹⁷L. D. Jackel, W. W. Webb, J. E. Lukens, and S. S. Pei, Phys. Rev. B 9, 115 (1974).
- ¹⁸R. De Luca, S. Pace, and A. Saggese, IEEE Trans. Appl. Supercond. 3, 1402 (1993).
- ¹⁹V. Calzona, M. R. Cimberle, C. Ferdeghini, F. Pupella, M. Putti, C. Rizzuto, A. Siri, and R. Vaccarone, Cryogenics **30**, 569 (1990).
- ²⁰S. Pace, A. Saggese, R. De Luca, F. Celani, and L. Liberatori, IEEE Trans. Magn. 27, 1049 (1991).
- ²¹M. R. Beasley, R. Labusch, and W. W. Webb, Phys. Rev. 181, 682 (1969).
- ²²B. Oh, R. H. Koch, and W. J. Gallagher, Phys. Rev. B 46, 9169 (1992).
- ²³M. Majoroš, P. Kottman, M. Polák, W. Schauer, V. Windte, J. Reiner, and V. Plecháček, Cryogenics **32**, 1061 (1992).
- ²⁴H. G. Schnack, R. Griessen, J. G. Lensink, C. J. van der Beek, and P. H. Kes, Physica C 197, 337 (1992).