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Generalized inversion scheme for the determination of activation energies from flux-creep experiments in high-T_c superconductors

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For thermally activated flux creep, which can be characterized by a temperature-, magnetic-field-, and current-density-dependent activation energy $U(j,T;B_e)$, we show that this function can be determined from a combination of magnetization and magnetization dynamical relaxation rate data. As an illustration the method is applied to creep data on epitaxial $YBa_2Cu_3O_7$ thin films. In contrast to previously proposed procedures, the present inversion scheme is much more general as it does not require *a priori* assumptions about the explicit temperature or field dependence of U.

Since the discovery by Müller, Takashige, and Bednorz[1] of large and nonexponential relaxation of the magnetic moment of $(La,Ba)_2CuO_{4-y}$ ceramics, giant flux creep has been found and investigated in many high-T_c superconductors. Most of the theories published recently (see, e.g., Refs. 2–7) explain this giant flux creep in terms of thermally activated flux motion (TAFM) (except at low temperature, typically below 1 K, where tunneling of flux lines seems to be the dominant dissipation mechanism[8]). The central parameter of TAFM theories is the energy barrier which has to be thermally overcome by flux lines or bundles of flux lines to move from one flux line configuration to another. The energy barrier depends on temperature T, magnetic induction B, and current density j. It depends furthermore on the direction of movement of the flux line bundle. In this paper we shall only consider thermally activated hopping *in* the direction of the Lorentz force acting on a flux line. Data taken above the irreversibility line will not be considered since they are strongly influenced by flux line hopping in a direction *opposite* to the Lorentz force. The differential equation governing thermally activated flux creep is[9]

$$\frac{\partial \boldsymbol{B}}{\partial t} = -\nabla \times \left[\frac{\boldsymbol{B} \times (\boldsymbol{j} \times \boldsymbol{B})}{|\boldsymbol{B} \times \boldsymbol{j}|} x_0 \omega_0 e^{-U(j,T,B)/kT} \right], \quad (1)$$

where $\boldsymbol{B}$ is the magnetic induction obtained by averaging the local field inside the superconductor over several penetration depths, $\boldsymbol{j}$ is the supercurrent density, $2x_0$ the hopping distance, and ω_0 the attempt frequency. In deriving Eq. (1) we have assumed that flux lines are moving perpendicularly to both $\boldsymbol{B}$ and $\boldsymbol{j}$, which is appropriate for high-κ superconductors.

For a sample with cylindrical symmetry in a magnetic field H_e parallel to the axis of symmetry Eq. (1) is considerably simplified, as $\boldsymbol{j} = (0, j, 0)$ in cylindrical coordinates (r, ϕ, z). After integration over the cross-sectional area we obtain

$$\frac{dj}{dt} = \frac{\chi_0}{\mu_0 \Omega} \frac{dB_e}{dt} - \frac{\Delta x_0 \omega_0 B_e}{\mu_0 \Omega} e^{-U(j,T,B_e)/kT}, \quad (2)$$

where χ_0 is the differential susceptibility, Δ a geometric factor, Ω the proportionality factor between j and the magnetic moment of the sample, and $B_e = \mu_0 H_e$. In Eq. (2) we have taken the averaged current density over the sample, and replaced B by B_e in the activation energy, which is a good approximation as tested by Griessen *et al.*,[10] Schnack *et al.*,[11] and Van der Beek *et al.*[12] (we consider here only data for fully penetrated samples). In the last reference it is also shown that the geometry of the sample plays a minor role in the determination of activation energies. For an infinitely long cylinder, $\chi_0 = \pi R^2$, $\Delta = 2\pi R$, $\Omega = \pi R^3/3$, where R is the radius of the sample, and for a thin sample, $\chi_0 = \pi^2 R^3/(3\mathcal{L})$, $\Delta = 2\pi^2 R^2/(3\mathcal{L})$, $\Omega = \pi R^3 D/3$, where $D \ll R$ is the thickness of the sample, and $\mu_0 R\mathcal{L}$ its self-inductance, which is of the order of $\ln(R/D)$. Feigel'man *et al.*[13] proposed the so-called interpolation formula for the current-dependent activation energy $U(j) = (U_c/\mu)((j_c/j)^\mu - 1)$ for collective creep in a system with randomly distributed weak pinning centers. The values of the exponent μ range from 1/7 to 3/2, depending on the current and temperature regime and the dimensionality of the system. For the simple Kim-Anderson model $\mu = -1$ and for Zeldov *et al.*'s logarithmic potential[14] $\mu = 0$. Each of these models gives a specific relaxation behavior that can be compared with experimental results. However, it is not possible to derive $U(j,T;B_e)$ directly from the experimental data without making specific assumptions about the explicit temperature and field dependence of U.

In this paper we take a different approach and show that the activation energy $U(j,T;B_e)$ can be determined from relaxation *and* current measurements. In our analysis the current and temperature dependences of the activation energy are written as

$$U(j,T;B_e) = g(T;B_e) f(J), \quad (3)$$

where $J = J(T;B_e) = j/j_c(T;B_e)$; j_c is the true critical current, for which the activation energy vanishes [i.e., $U(j_c(T;B_e),T;B_e) = 0$, and consequently $f(1) = 0$], otherwise f and g are arbitrary differentiable functions. Since only the product $g \times f$ is a physically observable quantity we can arbitrarily take $g(0;B_e) = 1$. The problem has now been reduced to the determination of $f(J)$ and $g(T;B_e)$.

In a magnetization hysteresis loop (MHL) measurement, i.e., when the external field is continuously swept, we have in most cases $|dj/dt| \ll (\chi_0/\mu_0\Omega)dB_e/dt$ so that the flux creep equation (2) can be rewritten as[11]

$$U(j_s, T; B_e) = CkT, \tag{4}$$

with $C \equiv \ln[2x_0\omega_0 B_e/a(dB_e/dt)]$. Equation (4) which defines implicitly the current j_s setup in the sample during a field sweep shows that j_s depends on T, dB_e/dt, and B_e, i.e., $j_s = j_s(T, B_e, dB_e/dt)$. Note that j_s is always smaller than the true critical current j_c, as long as we are in the creep regime. By substituting Eq. (3) into Eq. (4) and taking the *total* derivative with respect to T, we find

$$1 = \frac{d\ln U}{d\ln T} = \frac{d\ln g}{d\ln T} + \frac{d\ln f}{d\ln J} \times \frac{d\ln J_s}{d\ln T}, \tag{5}$$

with $\ln J_s = \ln j_s - \ln j_c$. Similarly by differentiation of Eq. (4) with respect to dB_e/dt at constant T, we obtain

$$\frac{d\ln f}{d\ln J} \times \frac{d\ln J_s}{d\ln[dB_e/dt]} = \frac{d\ln C}{d\ln[dB_e/dt]} = -\frac{1}{C}. \tag{6}$$

By noting that $j_c(T; B_e)$ does not depend on dB_e/dt, substitution of Eq. (6) into Eq. (5) gives us

$$1 = \left[\frac{d\ln j_c}{d\ln T} - \frac{d\ln j_s}{d\ln T}\right] / \left[C\frac{d\ln j_s}{d\ln[dB_e/dt]}\right] + \frac{d\ln g}{d\ln T}, \tag{7}$$

where $Q \equiv d\ln j_s/d\ln[dB_e/dt]$, the dynamical relaxation rate[11,15], can be determined from measurements of the dependence of the width of the MHL (or of the current j_s) on the field sweep rate at constant temperature. $d\ln j_s/d\ln T$ can be measured by taking the width of the MHL at a constant dB_e/dt, for different temperatures. C can also be calculated from the measured data since at low temperatures g and j_c hardly change with temperature. Setting these derivatives equal to zero, one obtains from Eq. (7)

$$C = \lim_{T\to 0} -\frac{1}{Q}\frac{d\ln j_s}{d\ln T}. \tag{8}$$

Since most of the terms in Eq. (7) are accessible to experiment, Eq. (7) is thus a relation between $d\ln g/d\ln T$ and $d\ln j_c/d\ln T$. At this point the usual procedure[3–5] is to postulate an *a priori* temperature dependence of $g(T; B_e)$ and calculate the corresponding $j_c(T; B_e)$.

There is, however, a much more general way to proceed, which is based on the fact that for a wide class of flux pinning models the characteristic pinning energy U_c is essentially proportional to j_c, i.e., $U_c(T; B_e) \propto [j_c(T; B_e)]^p$. As our function $g(T; B_e)$ can be associated with $U_c(T; B_e)$ this implies that $d\ln g/d\ln T = d\ln U_c/d\ln T = p(d\ln j_c/d\ln T)$. The exponent p depends on the pinning regime and the dimensionality of the system. For a collectively pinned single vortex in three dimensions (3D) $p = 1/2$ since $U_c \approx j_c\Phi_0 L_c\xi$ where Φ_0 is the flux quantum, ξ the coherence length, and $L_c \propto j_c^{-1/2}$ is the correlation length parallel to the field.[13] In 2D, in the same regime, $p = 1$ since L_c is limited by the thickness of the layers. At sufficiently high fields, flux lines will move in bundles. Within the framework of theoretical collective creep models,[13,16,17] in 3D systems the pinning barrier for a flux bundle is $U_c \approx B_e j_c V_B r_p$, where $V_B = R_\parallel R_\perp L_c$ is the volume of the bundle, with $R_\parallel$ and $R_\perp$ the dimensions of the bundle parallel and perpendicular to the displacement, respectively, and L_c the length of the bundle in the direction of B_e. The range of the potential r_p depends on temperature[18] as $r_p \approx \xi(1 + T/T_L)^{1/2}$, with T_L a characteristic temperature. The energies of elastic deformation of the flux line lattice should be of the same order of magnitude for all kinds of deformations:

$$V_B C_{11}(r_p/R_\parallel)^2 \sim V_B C_{66}(r_p/R_\perp)^2 \sim V_B C_{44}(r_p/L_c)^2 \sim U_c. \tag{9}$$

The elasticity moduli C_{ii} can be expressed in terms of B_e, Φ_0, penetration depth λ, and correlation length[16] R_c. From Eq. (9) and the expressions for C_{ii} given in Ref. 16 one finds, for example,

$$U_c(T) j_c^{3/2}(T) \approx A\frac{\xi(T)^{7/2}}{\lambda_\parallel(T)^4\lambda_\perp(T)}\left(1 + \frac{T}{T_L}\right)^{7/4}, \tag{10}$$

with $A = (B_e^2/\mu_0)[\Phi_0/(16\pi\mu_0)]^{3/2}$, for the nonlocal limit (i.e., $R_c \ll \lambda_\perp$). For the local limit we find a similar formula, but with a different exponent for j_c: 1/2. The temperature dependence of the quantities on the right-hand side of Eq. (10) partly cancel each other, resulting in a temperature dependence of the product $U_c(T) j_c^{3/2}(T)$ that is weaker than that of U_c and j_c taken separately.[16] For the 2D case[17] one finds a similar result, with $p = -1/2$ in the nonlocal limit, and $p = 0$ in the local limit. Thus, for simplicity we write[19]

$$g(T; B_e) \propto U_c(T; B_e) \propto [j_c(T; B_e)]^p, \tag{11}$$

with $p = -3/2, -1/2, 0, 1/2$, or 1. In the following p is treated as a free parameter.

Having a relation between $g(T; B_e)$ and $j_c(T; B_e)$ at hand, we can insert it in Eq. (7), and calculate the true critical current, for a given B_e,

$$j_c(T) = j_s(T)\exp\left[\int_0^T CQ(T')\frac{1 - p\frac{d\ln j_s}{d\ln T'}}{1 + pCQ(T')}\frac{dT'}{T'}\right] \tag{12}$$

as a function of T. Similarly for the explicit temperature dependence of the activation energy we find:

$$g(T) = \exp\left[\int_0^T p\frac{CQ(T') + \frac{d\ln j_s}{d\ln T'}}{1 + pCQ(T')}\frac{dT'}{T'}\right]. \tag{13}$$

Finally, with the help of Eqs. (3) and (4), we can construct $f(J)$ point by point:

$$f(J_s(T)) = CkT/g(T), \tag{14}$$

where T is playing the role of a running parameter and $J_s(T) = j_s(T)/j_c(T)$.

Equations (12) – (14) are the central formulas of our

inversion scheme. So far we have only described the generalized inversion scheme for dynamical relaxation data (using j_s and Q as input data). It is, however, completely straightforward to formulate the inversion scheme for conventional relaxation data. Equation (4) must then simply be replaced by $U(j(t),T;B_e) = kT\ln[(\tau_i+t)/\tau]$ as discussed in Refs. 11 and 15. Furthermore, also current-voltage (I-V) measurements can be included in our data set, as $I \propto j_s$ and $V \propto E \propto dB_e/dt$, where E is the electric field.[11] These type of data provide us with valuable extra information, especially when the current extends over several decades. The electric field E changes then over many decades. This corresponds to a considerable variation of C and, according to Eq. (4), of U. As T is constant for a single I-V curve, and $g(T;B_e)$ is known from the inversion scheme, each I-V curve is in fact a scaled f vs J curve. Below, when we apply our inversion scheme to a specific set of experimental data, we show that the combination of dynamical relaxation data and I-V data provides us with a method to determine p.

At this point it is, however, instructive to compare the present inversion scheme with various data analysis methods used so far. Most of them are just special cases of our inversion scheme. For example, Maley *et al.*[2] assume that both j_c and g are temperature independent. Equation (7) reduces then to Eq. (8) for all temperatures and the function f can be constructed directly. In fact most methods use Eq. (7) too, and try to keep C constant, eventually by choosing explicitly $j_c(T)$ and $U_c(T)$. McHenry *et al.*[3] assume a definite temperature dependence $g(T) = 1-(T/T^*)^2$ and keep j_c constant. This corresponds to $p = \infty$ in our formulas. Then they determine T^* by requiring that C remains approximately constant in Eq. (13) with $p = \infty$. Similarly Thompson *et al.*[6] assume that U_c and j_c depend in the same way on $1-(T/T_c)^2$, and thus $p = 1$. The choice of a quadratic T dependence is in fact not necessary since $g(T)$ can be constructed point by point from Eq. (13) (with $p = \infty$ or $p = 1$). Sengupta *et al.*[7] take only j_c as a function of T, which can be described with $p = 0$ in our scheme.

As an illustration we apply our generalized inversion scheme to magnetization measurements on an $YBa_2Cu_3O_7$ epitaxial film of 180 nm thickness.[20] The input data for the inversion scheme are shown in Fig. 1. They consist of $j_s(T)$ and $Q(T)$ data at sixteen temperatures, and I-V curves at five temperatures. For all data $B_e = 0.5$ T. The saturation of j_s and Q at low temperatures is clearly not compatible with a pure TAFM model, but is a manifestation of (thermally assisted) quantum flux creep. Since our inversion scheme presupposes thermally activated flux jumps we need to estimate $j_s^*(T)$ and $Q^*(T)$ corresponding to $j_s(T)$ and $Q(T)$, respectively, in the absence of quantum creep. At low temperatures ($T < 13$ K) $j_s^*(T)$ is obtained from an extrapolation of j_s data at $T > 13$ K. This extrapolation leads to $j_s^*(0) = j_c(0) = 8.2\times10^{10}$ A/m^2. Since $Q^*(0) = 0$ in any TAFM model the low-temperature behavior of $Q^*(T)$ can easily be obtained from an interpolation between 0 at 0 K and data at $T > 13$ K. For the parameter C an extrapolation to 0 K of $(-1/Q)d\ln j_s/d\ln T$ of data for $T > 13$ K leads also to a good estimate ($C = 14$). The accuracy

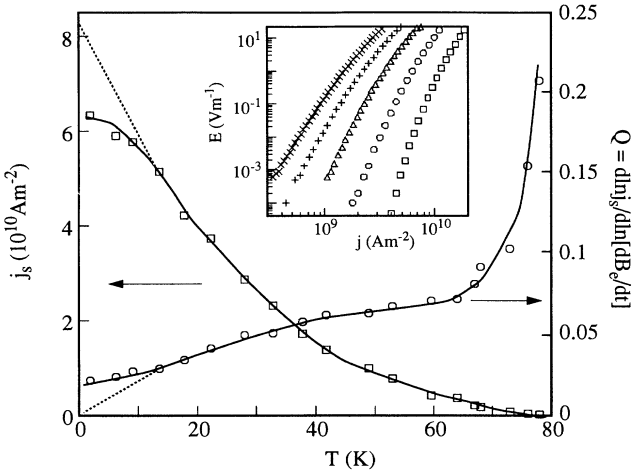

FIG. 1. Experimental results at $B_e = 0.5$ T for an $YBa_2Cu_3O_7$ epitaxial film of 180 nm thickness, used as input data for the generalized inversion scheme. The current j_s in the sample is determined from magnetic moment measurements at a constant sweep rate of 0.04 T/s, and I-V curves at high temperatures (inset). The dynamical relaxation rate Q is determined from MHL measured at various sweep rates. The data points at higher temperatures are obtained from the slope of current-voltage curves on the same film taken at an electric field $E = 7.5\times10^{-5}$ V/m, corresponding to the 0.04 T/s sweep rate. The dashed lines indicate $j_s^*(T)$ and $Q^*(T)$ corresponding to $j_s(T)$ and $Q(T)$ in the absence of quantum creep.

of the extrapolated values is not critical since at low temperatures the integrand in Eq. (13) vanishes and Eq. (12) reduces to $j_c(T) \approx j_c(0)$. Application of Eqs. (12)–(14) to the data leads to the results shown in Fig. 2, where U is plotted as a function of j and T for $C = 14$ and $p = 0.65$. This value of p which is close to $p = 1/2$ predicted for

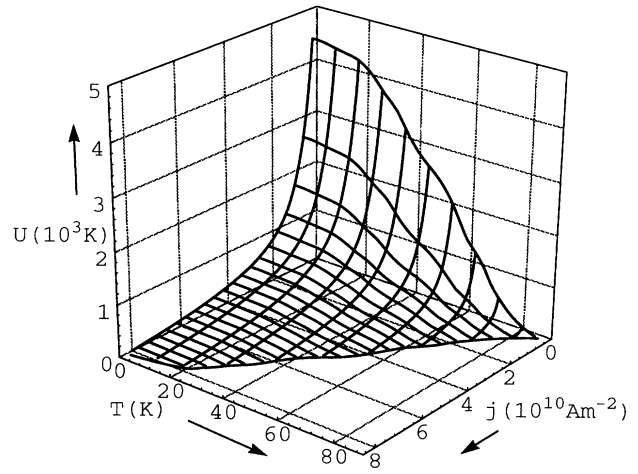

FIG. 2. A three-dimensional plot of $U(j,T;B_e = 0.5$ T) for the data of Fig. 1, obtained from the inversion scheme [Eqs. (12)–(14)] with $p = 0.65$ and $C = 14$. Note that the temperature dependence of constant-j lines is not simply given by the function $g(T)$ since the argument of the function f in Eq. (3) depends also on T via $j_c(T)$.

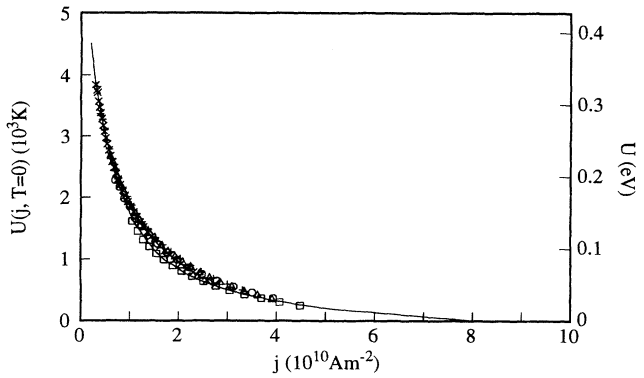

FIG. 3. Activation energy $U(j, T = 0)$ obtained from the generalized inversion scheme (see Fig. 2) (line) together with $U(j, T = 0)$ derived from the I-V curves in the inset of Fig. 1, by using the relation $E = v_0 B_e \exp[-U(j, T; B_e)/kT]$, the value of v_0 corresponding to $C = 14$ and the functions $g(T)$ and $j_c(T)$ determined from the inversion scheme. N.B. in the inversion scheme only the slope of the I-V curves at $E = 7.5 \times 10^{-5}$ V/m are used as input data. The data points, however, correspond to the whole I-V curve. The symbols are the same as in Fig. 1.

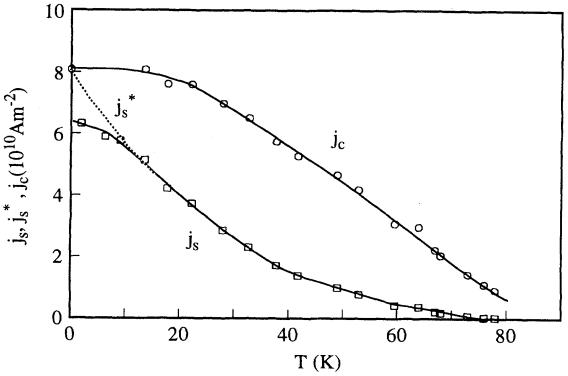

FIG. 4. True critical current $j_c(T)$ calculated for $p = 0.65$ and $C = 14$, by means of the inversion scheme [Eq. (12)], for the data of Fig. 1. The measured current j_s is shown for comparison. The dashed line corresponds to $j_s^*(T)$, the current in the absence of quantum creep.

a collectively pinned vortex in a 3D system, has been determined from the requirement that the activation energy determined from the five I-V curves shown in the inset of Fig. 1 fall as well as possible on the $U(j, T = 0)$ curve obtained from the inversion of $j_s(T)$ and $Q(T)$ data only (i.e., data taken at $dB_e/dt = 0.04$ T/s which corresponds to an electric field of 7.5×10^{-5} V/m). The very good overlap shown in Fig. 3 demonstrates clearly that it is possible to describe TAFM over a wide temperature and current range by means of a unique activation energy function. In Fig. 4 the corresponding $j_c(T)$ is shown, together with the measured $j_s(T)$. At all temperatures j_s is lower than j_c. The difference between j_c and j_s is of course larger at higher temperatures.

In a forthcoming publication[20] the generalized inversion scheme shall be applied to data for $YBa_2Cu_3O_7$ and $YBa_2Cu_4O_8$ films over the same temperature range and for fields up to 7 T. In that work we shall also show that even if data near the irreversibility line are not available, an estimate for p can nevertheless be found.

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