Distribution of Activation Energies for Thermally Activated Flux Motion in High-T_c Superconductors: An Inversion Scheme

C. W. Hagen and R. Griessen

Natuurkundig Laboratorium, Vrije Universiteit, De Boelelaan 1081, 1081 HV Amsterdam, The Netherlands

(Received 10 February 1989)

Within a thermally activated flux-motion model we derive an exact inversion scheme which makes it possible to calculate the distribution $m(E^*)$ of activation energies E^* for flux motion from experimental magnetic relaxation data $M(t,T)$. The distributions determined from relaxation data for polycrystalline and single-crystalline $YBa_2Cu_3O_{7-\delta}$ strongly resemble a log-normal distribution function. The results show that within this model existing data imply that structural disorder is present in both ceramic and single-crystalline samples.

PACS numbers: 74.60.Ge, 74.70.Vy

Soon after the discovery of high-T_c superconductivity, Müller, Takashige, and Bednorz[1] reported on nonexponential time decay of the magnetization M of these superconductors. These observations,[2] together with the presence of an irreversibility line which showed a close resemblance with the de Almeidia-Thouless line in spin glasses, led to the assumption of a glassy state in high-T_c superconductors and to the suggestion[1,3] that $M(t) \propto \exp[-(t/\tau)^{\beta}]$, i.e., Kohlrausch behavior. This idea appeared quite reasonable for ceramic samples consisting of weakly linked superconducting grains. Later, however, large relaxation effects were reported also for a single crystal[4] of $YBa_2Cu_3O_{7-\delta}$. Many authors, among which are Rossel and Chaudhari,[5] Hagen *et al.*,[6] Yeshurun and Malozemoff,[4] and Tinkham,[7] pointed out that these relaxation effects might also rise from thermally activated flux motion (TAFM), which was known to occur in conventional superconductors, though on a much smaller scale. Anderson[8] and Beasley, Labusch, and Webb[9] showed that in the limit where the activation energy E for TAFM is much larger than kT, the magnetic relaxation follows a logarithmic law. As a Kohlrausch behavior with $\beta \ll 1$ is difficult to distinguish from a logarithmic time dependence, it has not been possible yet to identify unambiguously the origin of the large magnetic relaxation effects observed in all high-T_c superconductors. Recent measurements by Rossel, Maeno, and Morgenstern[10] of memory effects in the magnetic relaxation seems to indicate that glassy behavior occurs only in relatively weak magnetic fields ($B \sim 0.2$ T) at temperatures close to T_c. For a discussion of the validity of the superconducting-glass or flux-motion picture the reader is referred to Refs. 11–14. In this Letter we shall analyze magnetic relaxation data within a TAFM model and show that the distribution of activation energies can be determined unambiguously from the temperature dependence of the magnetic relaxation.

We focus our attention on the temperature dependence of the magnetic relaxation. In Fig. 1 we show the results obtained by Tuominen, Goldman, and Mecartney[15,16] for both polycrystalline and single-crystalline $YBa_2Cu_3O_{7-\delta}$, by Rossel and Chaudhari[5] for an epitaxial film of $YBa_2Cu_3O_{7-\delta}$ on (100) $SrTiO_3$, and by Yeshurun, Malozemoff, and Holtzberg[13] for a single crystal. For three samples the normalized logarithmic derivative $-d\ln M/d\ln t$ exhibits a peak as a function of temperature. As discussed by various authors[17,18] the temperature variation exhibited in Fig. 1 cannot be understood in terms of TAFM over barriers with a constant height, as then

$$-\left.\frac{d\ln M}{d\ln t}\right|_{t_b} = \left(\frac{E(T)}{kT} - \ln\frac{t_b}{\tau}\right)^{-1}, \qquad (1)$$

where t_b is the time at which magnetic relaxation data are recorded and τ are a relaxation time which we shall see is typically 10^{-10} to 10^{-12} s. As the activation energy $E(T)$ decreases to zero at $T = T_c$, Eq. (1) implies that at a certain temperature $T_d < T_c$, $d\ln M/d\ln t$ should diverge, in contradiction with the experimental data in Fig. 1.

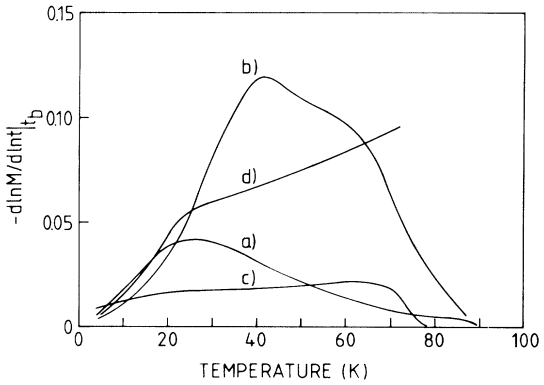

FIG. 1. Temperature dependence of the relative relaxation rate $(d\ln M/d\ln t)_{t=t_b}$ for, curve *a*, polycrystalline bulk $YBa_2Cu_3O_{7-\delta}$ (Ref. 15) with $t_b = 600$ s; curve *b*, single-crystalline $YBa_2Cu_3O_{7-\delta}$ (Ref. 16) with $t_b = 60$ s; curve *c*, epitaxial $YBa_2Cu_3O_{7-\delta}$ film on (100) $SrTiO_3$ (Ref. 5) with $t_b = 150$ s; and curve *d*, single crystal (Ref. 13) with $t_b = 200$ s.

Within a TAFM model it is thus necessary to postulate a distribution of activation energies to explain the relaxation data. We consider a superconductor consisting of a collection of regions with different activation energies. Within a given region the pinning centers are separated from each other by barriers of height E^*. Each pinning center can accommodate several flux lines. The distribution of activation energies $m(E^*)$ is defined in such a way that $m(E^*)$ is the fraction of barriers with activation energies between E^* and E^*+dE^* at $T=0$. Then the total magnetic moment $M(t,T)$ at time t and temperature T may be written as

$$M(t,T)=M_0\frac{b(T)}{a(T)}\int_{E_0^*(t,T)}^{\infty}m(E^*)\left[1-\frac{kT}{E^*b(T)}\ln\left(1+\frac{t}{\tau}\right)\right]dE^* \tag{2}$$

since, as shown in Refs. 19 and 20, the magnetic relaxation of a process with an activation energy E is given by an expression of the form $M(t)=M_0[1-(kT/E)\ln(1+t/\tau)]$ even in the case where E is comparable to kT. In Eq. (2), $E_0^*(t,T)\equiv b^{-1}(T)[kT\ln(1+t/\tau)]$. The function $b(T)$ describes the temperature dependence of the activation energy and is by definition equal to 1 at $T=0$, i.e., $b(0)=1$. The ratio $b(T)/a(T)$ is equal to $j_m(T)/j_m(0)$, where $j_m(T)$ is the maximum current which is compatible with a thermally activated model. In other words, $j_m(T)$ would be equal to the measured critical current density $j_c(T)$ if thermal fluctuations (hopping of flux lines) did not occur. In Eq. (2) the distribution function $m(E^*)$ depends only on E^* and not on T as it is by definition the distribution of activation energies at *zero* temperature. We require that it is normalized so that

$$\int_0^{\infty}m(E^*)dE^*=1. \tag{3}$$

As for all experimental studies carried out so far, $t\gg\tau$, we replace in the following $\ln(1+t/\tau)$ by $\ln(t/\tau)$. At this point one might select a certain expression for the distribution function $m(E^*)$, calculate $M(t,T)$, and compare the results to experimental data as was done by Malozemoff *et al.*[17] However, it would be much more preferable to find a procedure to determine uniquely $m(E^*)$ from the experimental data.

For this we consider the derivative $dM/d\ln t$ at the time of the experiment t_b. This derivative is given by

$$\frac{dM}{d\ln t}=-M_0\frac{kT}{a(T)}\int_{E_0^*(t_b,T)}^{\infty}\frac{m(E^*)}{E^*}dE^*. \tag{4}$$

By differentiating Eq. (4) with respect to T we obtain that

$$m(E_0^*(t_b,T))=\left[\frac{d}{dT}\left(\frac{a(T)}{M_0kT}\frac{dM}{d\ln t}\right)\right]\left[\frac{b(T)}{T}\frac{d}{dT}\left(\frac{T}{b(T)}\right)\right]^{-1}. \tag{5}$$

As E_0^* depends on the *a priori* unknown relaxation time τ, Eq. (5) is not yet suitable for an inversion of experimental data. It is, however, possible to derive another expression for $m(E_0^*(t_b,T))$. Consider Eq. (2) which can be rewritten as follows, by using Eq. (4):

$$M(t,T)=M_0\frac{b(T)}{a(T)}\int_{E_0^*(t,T)}^{\infty}m(E^*)dE^*+\ln\left(\frac{t}{\tau}\right)\frac{dM}{d\ln t}. \tag{6}$$

By differentiating with respect to temperature we obtain

$$m(E_0^*(t_b,T))=\left\{\frac{d}{dT}\left\{\frac{a(T)}{M_0b(T)}\left[\ln\left(\frac{t_b}{\tau}\right)\left(\frac{dM}{d\ln t}\right)-M\right]\right\}\right\}\left[\ln\left(\frac{t_b}{\tau}\right)\frac{d}{dT}\left(\frac{kT}{b(T)}\right)\right]^{-1}. \tag{7}$$

Dividing Eq. (7) by Eq. (5) and rearranging terms we find

$$\ln\left(\frac{t_b}{\tau}\right)=\left[T\frac{b(T)}{a(T)}\frac{d}{dT}\left(\frac{M(t_b,T)}{b(T)}a(T)\right)\right]\left[\left(\frac{dM}{d\ln t}\right)\left(1-\frac{d\ln b(T)}{d\ln T}\right)\right]^{-1}. \tag{8}$$

Equation (5) or (7) together with Eq. (8) make it possible to invert experimental data for $M(t_b,T)$ into the distribution function $m(E^*)$ when the temperature dependence of the activation energies $[b(T)]$ and of the Lorentz term force $[a(T)]$ are known. At lower temperatures $a(0)=1$ and $b(0)=1$ so that $\ln(t_b/\tau)$ can directly be determined from the measured quantities $M(t_b,T)$ and $dM/d\ln t$. As τ is assumed to be temperature independent, Eq. (8) represents then a condition for the temperature dependence of $a(T)$ and $b(T)$. As the activation energy is proportional to the condensation energy in a correlated volume V, $b(T)$ is assumed to have the following temperature dependence $b(\Theta)=(1-\Theta^2)^2[(1$

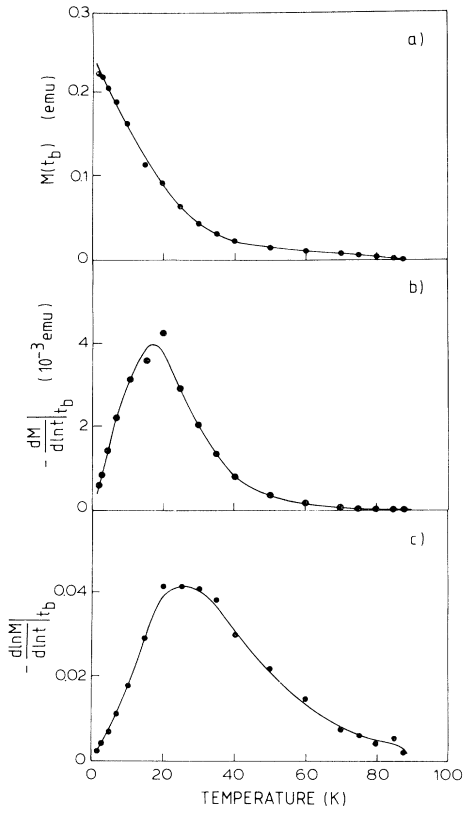

FIG. 2. Temperature dependence of (a) $M(t_b,T)$, (b) $dM/d\ln t$, and (c) $d\ln M/d\ln t$ for the polycrystalline $YBa_2Cu_3O_{7-\delta}$ sample from Ref. 15. The experimental time is $t_b=600$ s. The solid lines are fits to the experimental data and are used in the inversion scheme.

$+\Theta^2)/(1-\Theta^2)]^{n/2}$ with $\Theta=T/T_c$ and $0\leq n\leq 3$ as $Eb(\Theta)$ is proportional to $B_c^2(\Theta)\xi^n(\Theta)$. The last term arises from the temperature dependence of the coherence length[21] which follows from the empirical dependences $B_c=B_c(0)(1-\Theta^2)$ and $\lambda^2=\lambda^2(0)/(1-\Theta^4)$. The model recently proposed by Tinkham,[22] where $E\propto B_c^2 a_0^2\xi$ with $a_0^2=\phi_0/B$ (ϕ_0 is the flux quantum), corresponds to the choice $n=1$.

For the function $a(T)$ one expects that it varies as $VL_{\rm hop}$, where $L_{\rm hop}$ is the distance over which flux lines jump from one pinning region to the other, i.e., $a(\Theta)=[(1+\Theta^2)/(1-\Theta^2)]^{m/2}$ with $0\leq m\leq 4$. The case $m=0$ corresponds to V and $L_{\rm hop}$ independent of the coherence length, i.e., to a case where the microstructure of the sample itself is the determinant factor. In contrast $m=4$ corresponds to the case where all relevant lengths are related to the coherence length.

Both for the polycrystalline[15] and the single-crystalline data[13] Eq. (8) is best satisfied with $n=2$ and $m=0$. All other combinations of the parameters n and m lead to a strong temperature dependence of τ in disagreement with our assumption.

We consider now the results obtained by means of the inversion scheme described above for two sets of data.

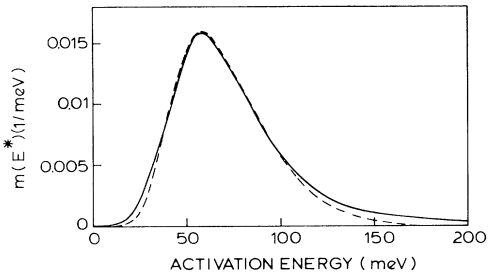

FIG. 3. Distribution function of activation energies (solid line) as obtained from inverting the data of Ref. 15 for a polycrystalline $YBa_2Cu_3O_{7-\delta}$ sample. The shape of the distribution function resembles a log-normal distribution, shown as the dashed line, for $P_m=0.016$, $E_m^*=60$ meV, and $\gamma=4$ (see text).

(a) Polycrystalline $YBa_2Cu_3O_{7-\delta}$ sample.—The temperature dependence of $M(t_b,T)$ and its time derivatives determined from the data of Tuominen, Goldman, and Mecartney[15] are shown in Fig. 2. From these data we determined $\ln(t_b/\tau)=30.4$. By means of Eq. (8) with $t_b=600$ s, this implies that $\tau\sim 4\times 10^{-11}$ s, a value comparable to a typical phonon frequency. By means of Eq. (5) or (7) and the data in Fig. 2 we determined then the distribution function $m(E^*)$ shown in Fig. 3.

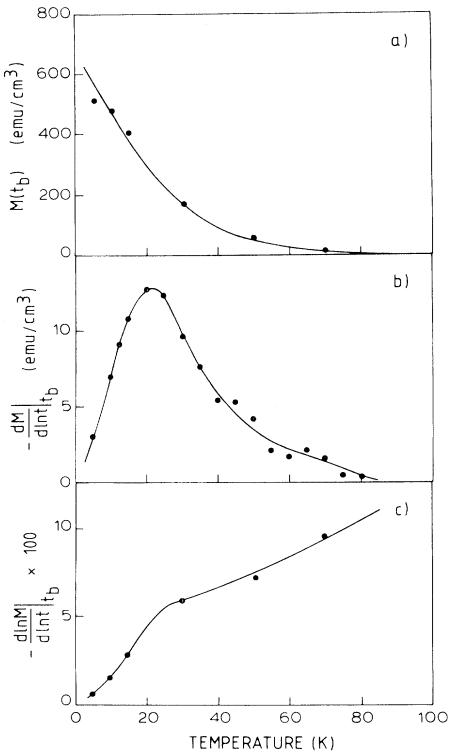

FIG. 4. Variation with temperature of (a) $M(t_b,T)$, (b) $dM/d\ln t$, and (c) $d\ln M/d\ln t$ for the single crystal of Ref. 13. The solid lines fitted to the data are used for the inversion of the relaxation data into an activation energy distribution $m(E^*)$.

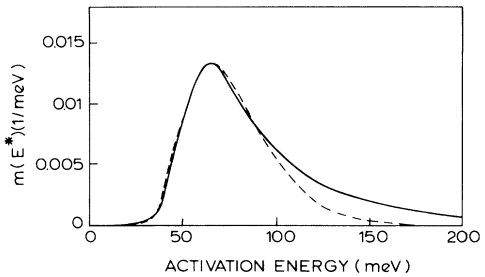

FIG. 5. Distribution function (solid line) for a single crystal (Ref. 13) of $YBa_2Cu_3O_{7-\delta}$. The dashed line is a log-normal distribution with $P_m=0.0134$, $E_m^*=67$ meV, and $\gamma=5.52$ (see text).

(b) Single crystal of $YBa_2Cu_3O_{7-\delta}$.—From the data of Yeshurun, Malozemoff, and Holtzberg[13] on $dM/d\ln t$ and the $M(t,T)/M(t_b,T)$ data with $t_b=200$ s (Figs. 1 and 2 in Ref. 13) we determined $M(t_b,T)$ and $d\ln M/d\ln t$ as shown in Fig. 4. From these data we obtained by means of our inversion scheme $\tau=8\times10^{-11}$ s and the distribution function shown in Fig. 5.

For both types of samples, $m(E^*)$ remarkably resembles a log-normal distribution function $P(E^*)=P_m\times\exp\{-\gamma[\ln(E^*/E_m^*)]^2\}$, where $P_m=P(E_m^*)$ and E_m^* is the energy corresponding to the maximum value of $P(E^*)$. The fit, which involves only the parameter γ as P_m and E_m^* are entirely determined by the maximum of the distribution, is good except at energies above 100 meV. It is not clear at present what significance has to be attributed to this resemblance, but the fact that the log-normal distribution is often found, for example, to describe well the particle size distribution of powders[23] suggests that it might be related to a distribution of extended defects. Both for single-crystalline and for ceramic $YBa_2Cu_3O_{7-\delta}$ the distribution of activation energies $m(E^*)$ has a width of typically 50 meV. This suggests that the activation barriers relevant for TAFM are not related to the size of the grains but to the internal structure of the grains.[24]

The presence of high-energy tails is consistent with the findings of Kapitulnik[25] and Palstra *et al.*,[26] who report relatively large values (400 meV to well above 1 eV) for activation energies determined from the analysis of *I-V* characteristics in the vicinity of T_c.

In conclusion, we have presented an inversion scheme for the conversion of magnetic relaxation data $M(t,T)$ to a distribution $m(E^*)$ of activation energies. The framework followed in this work is essentially that of a parallel decay model in which relaxation in each region occurs independently of that in other regions. More work is needed to decide whether or not hierarchical dynamical processes occur in $YBa_2Cu_3O_{7-\delta}$ or a genuine glassy state is required to explain the electrodynamical properties of high-T_c superconductors.[27]

[1]K. A. Müller, M. Takashige, and J. G. Bednorz, Phys. Rev. Lett. **58**, 1143 (1987).

[2]M. Takashige, K. A. Müller, and J. G. Bednorz, Jpn. J. Appl. Phys. Suppl. **26-3**, 1101 (1987).

[3]I. Morgenstern, K. A. Müller, and J. G. Bednorz, Z. Phys. B **59**, 33 (1987).

[4]Y. Yeshurun and A. P. Malozemoff, Phys. Rev. Lett. **60**, 2202 (1988).

[5]C. Rossel and P. Chaudhari, Physica (Amsterdam) **153C-155C**, 306 (1988).

[6]C. W. Hagen, E. Salomons, R. Griessen, and B. Dam (to be published).

[7]M. Tinkham, Helv. Phys. Acta **61**, 443 (1988).

[8]P. W. Anderson, Phys. Rev. Lett. **9**, 309 (1962).

[9]M. R. Beasley, R. Labusch, and W. W. Webb, Phys. Rev. **181**, 682 (1969).

[10]C. Rossel, Y. Maeno, and I. Morgenstern, Phys. Rev. Lett. **62**, 681 (1989).

[11]A. P. Malozemoff, Y. Yeshurun, L. Krusin-Elbaum, T. K. Worthington, D. C. Cronemeyer, T. Dinger, F. Holtzberg, T. R. McGuire, and P. H. Kes (to be published).

[12]A. P. Malozemoff, L. Krusin-Elbaum, D. C. Cronemeyer, Y. Yeshurun, and F. Holtzberg, Phys. Rev. B **38**, 6490 (1988).

[13]Y. Yeshurun, A. P. Malozemoff, and F. Holtzberg, in Proceedings of the Fourth Joint Magnetism and Magnetic Materials–Intermag Conference, Vancouver, 12–15 July 1988 (to be published).

[14]V. L. Aksenov and S. A. Sergeenkov, Physica (Amsterdam) **156C**, 235 (1988).

[15]M. Tuominen, A. M. Goldman, and M. L. Mecartney, Phys. Rev. B **37**, 548 (1988). [In their Fig. 3 the normalized rates of decay were erroneously plotted as $(-1/M_0)dM/d\log_{10}t$ instead of $(-1/M_0)dM/d\ln t$].

[16]M. Tuominen, A. M. Goldman, and M. L. Mecartney, Physica (Amsterdam) **153C-155C**, 324 (1988).

[17]A. P. Malozemoff, T. K. Worthington, R. M. Yandrofski, and Y. Yeshurun, in *Towards The Theoretical Understanding of High Temperature Superconductivity,* edited by S. Lundqvist, E. Tosatti, M. Tosi, and Y. Lu (World-Scientific, Singapore, 1988).

[18]M. Foldeaki, M. E. McHenry, and R. C. O'Handley, Phys. Rev. B (to be published).

[19]C. W. Hagen and R. Griessen, in "Thermally Activated Magnetic Relaxation in High-T_c Superconductors," edited by A. V. Narlikar (Nova Science Publishers, Commack, NY, to be published).

[20]C. W. Hagen, R. Griessen, and E. Salomons, Physica (Amsterdam) **157C**, 199 (1989).

[21]M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975).

[22]M. Tinkham, Phys. Rev. Lett. **61**, 1658 (1988).

[23]C. G. Granqvist and R. A. Buhrman, J. Appl. Phys. **47**, 2200 (1976).

[24]G. Deutscher and K. A. Müller, Phys. Rev. Lett. **59**, 1745 (1987).

[25]A. Kapitulnik (private communication).

[26]T. T. M. Palstra, B. Batlogg, L. F. Schneemeyer, and J. V. Wasczak, Phys. Rev. Lett. **61**, 1662 (1988).

[27]Jungzae Choi and Jorge V. José, Phys. Rev. Lett. **62**, 320 (1989).