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Magnetothermal instabilities in an organic superconductor

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Abstract. We have studied the occurrence of magnetothermal instabilities in a $\kappa-(\mathrm{BEDT-TTF})_2\mathrm{Cu[N(CN)_2]Br}$ single crystal during field sweep magnetization experiments, equivalent to short time relaxation studies. We find instability behaviour in good agreement with a recent model by Mints, for a non-linear $E(J)$ characteristic. In particular, we find that a decrease of the dynamic relaxation rate, characterizing the effective activation energy, precedes the unstable regime. We point out formal analogies between such instabilities and the general predictions for flux avalanches.

PACS. 74.60.Ge Flux pinning, flux creep, and flux-line lattice dynamics – 74.70.Kn Organic superconductors

Magnetothermal instabilities (flux jumps) from the critical state have been observed and modeled since the discovery of type-II superconductors with high critical currents (originally referred to as "hard superconductors") in the 1960s. The models which establish the stability criterion for the critical state all consider coupled electromagnetic and thermal equations. The flux jump is described as an amplification process, where a temperature rise induces a decrease of the critical current density, an entry of magnetic flux and an additional temperature increase. The earlier models assumed a linear current-voltage characteristic and two limiting cases were considered. The first one assumes that the flux diffusion is *adiabatic*, *i.e.* that the magnetic flux diffusion constant, D_M, is much larger than the thermal one [1–3]. The opposite case, which is referred to as the *dynamic approximation*, was considered later [4]. These modelations made clear that a way to eliminate flux jumps is to reduce the superconductor dimensions transverse to the magnetic field, as is done in multifilamentary wires. Many observations were made of a dependence of the occurrence of instabilities upon the field sweep rate, which could not be interpreted using a linear current-voltage characteristic [5]. Mints and Rakhmanov [6] showed, within the adiabatic approximation, that a non-linear characteristic could account for this observation. Recently, Mints [7] considered a logarithmic current-voltage characteristic, as obtained from several models for flux creep in the high current density limit. For parameter values typical of the high T_c materials, he finds that the dynamic approximation should be valid for magnetization measurements at a constant sweep rate and he derives the stability criterion in this case. Finally, Mints showed that the oscillations sometimes observed prior to the flux jumps can be deduced from the coupled electromagnetic and thermal equations [8]. The observation of flux jumps in the magnetization of some high T_c superconductors revived the interest for these early models. In his review of magnetic instabilities in high T_c superconductors [9], Wipf finds a general agreement between the adiabatic model and the data for $\mathrm{YBa_2Cu_3O_{7-\delta}}$ obtained by Tholence *et al.* [10], although he admits that the observed dependence of the occurrence of the instabilities with the field sweep rate appeals for some corrections. An extensive study of the thermal oscillations and jumps in a granular sample of $\mathrm{YBa_2Cu_3O_{7-\delta}}$ was given in [11]. These authors, in contrast to the approximations made by Mints [7], used a quadratic dissipation term in their analysis of their data (flux flow regime). As a consequence, the flux jumps and the oscillation periodicity are predicted to depend on different powers of the field sweep rate than the ones given in [7]. However, the data presented in reference [11] are either inconclusive or in contradiction with the predictions of the model given by the authors. As pointed out by Mints and Rakhmanov [5], very few experimental data lend themselves to a precise comparison with theory. The main reasons for this are generally uncontrolled perturbations during experiments and poor knowledge of the various material parameters involved. In this paper, we report on magnetization instabilities during field sweep on a single crystal of the organic superconductor $\kappa-(\mathrm{BEDT-TTF})_2\mathrm{Cu[N(CN)_2]Br}$. Our data exhibit well defined, reproducible flux jumps: two successive measurements under the same field sweep conditions give identical patterns of jumps. The torque technique that we have used allows for an almost continuous, quantitative registration of the magnetization, in contrast to SQUID magnetometer measurements. Also, we believe that this technique limits uncontrolled perturbations and scattering of the data. We show that the model by Mints accounts successfully for

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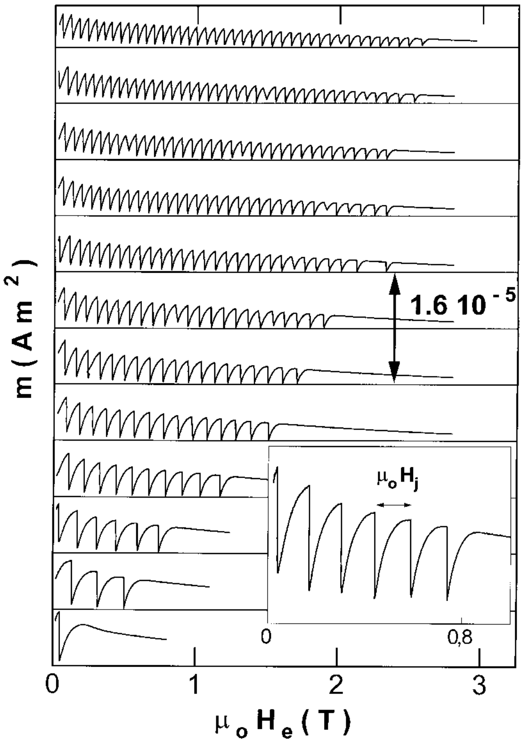

Fig. 1. Magnetic moment during field sweep experiments (low thermal coupling, $T_b = 1$ K). Bottom to top: $\mu_0 dH_e/dt = 0.06$, 0.1, 0.14, 0.18, 0.22, 0.26, 0.3, 0.34, 0.38, 0.42, 0.46 and 0.5 T/min. Insert: 0.14 T/min.

our data, and that an anomalous saturation and subsequent decrease of the magnetization with increasing field sweep rate observed at high rates may be understood within the same model.

The magnetization of the single crystal was measured using a capacitive torque experiment mounted on a dilution fridge. The temperature was measured directly on the metallic, highly conductive electrode which supports the sample. The magnetic field is applied at $\theta = 20°$ from the normal to the superconducting planes of the layered compound. Within the two dimensional approximation for this highly anisotropic material [12], the magnetic moment of the sample for a field H_e applied perpendicularly to the superconducting layers is:

$$m(H_e) = \Gamma(H_e/\cos\theta)/H_e \tan(\theta) \qquad (1)$$

where $\Gamma(H)$ is the torque for a magnetic field H applied at the angle θ. In the rest of the text, the magnetic moment displayed is the one given by this formula, *i.e.* the one for the *gedanken* experiment where the field is applied perpendicularly to the superconducting layers. Figure 1 shows the magnetization of a large single crystal $780 \times 660 \times 180$ μm^3 where the smallest dimension is the one transverse to the superconducting planes. In the following, we assimilate the sample to cylinder of radius R and length L, the exact geometry affecting the results by a numerical factor close to unity. In this first set of experiments, the sample was glued on its larger side to the capacitance electrode using a thick layer of vacuum grease (this set of experiments will be referred to as "low thermal coupling"). The magnetic moment exhibits periodic, well defined jumps to zero value when the magnetic field is swept at a constant rate. We have checked that the angle θ of the applied field has no influence on the magnetization deduced from the torque signal. A temperature rise of the metallic electrode is observed after each magnetization jump due to the sudden heat release related to the magnetization jump, but the nominal temperature is always recovered well before the following jump (Fig. 2). The sweep rate modifies the recovery $m(H_e)$ curve after a jump for a given field. In particular, it affects strongly the value of the field beyond which no instability subsists. A careful examination of the signal allows one to distinguish features in agreement with the magnetothermal instability model. First, the magnetization after a jump relaxes to zero (the "incomplete jumps" to a non zero value shown in Figure 1 are due to the finite sampling rate of the torque signal, $\mu_0 \Delta H_e = 1$ mT) in agreement with a temperature rise of the sample to a value where the screening current is negligible. Then, the jumps occur at a field at which the magnetization of the sample is still increasing with the applied field, showing that the sample has not reached a fully critical state. Finally, for fields just above the critical field at which no jumps are observed any longer, the magnetization tends to saturate to an almost field independent value, showing that the full critical state has been reached close to the vanishing of the instabilities. As shown by Mints [7], the sweep rate affects the instabilities due to the non-linear electric field characteristic $E(J)$. This is due to the increase of the differential resistivity, dE/dJ, and, hence, of the dissipated power, with increasing sweep rate. In a recent paper, he has determined the instability criterion in the case of a power law:

$$E = E_0(J/J_0)^n \qquad (2)$$

where E_0 and J_0 are some characteristic electric field and current density. In the case of a thermally activated resistivity $\rho = \rho_0 \exp(-U(J)/kT)$, where $U(J)$ is the activation energy, and in the limit of large current and large n, this is formally equivalent to a logarithmic dependence:

$$U(J) = U_0 \mathrm{Ln}(J_0/J), \quad n = U_0/kT \gg 1. \qquad (3)$$

As pointed out by Mints, the logarithmic form is known to be a good approximation for the more sophisticated dependencies inferred from various models, in the limit of large current densities. We shall use here such an approximation, as was done in [7]. Here, we should be clearly in the "dynamic" approximation where the magnetic flux diffusion constant related to vortex motion, D_M, is much smaller than the thermal one, D_T. Indeed, the magnetic flux diffusion constant may be expressed as:

$$D_M = \mu_0^{-1}\rho = \mu_0^{-1} dE/dJ \qquad (4)$$

(SI units, as for the following). The derivative dE/dJ during a field sweep experiment may be evaluated straightforwardly. The electric field during a field sweep experiment

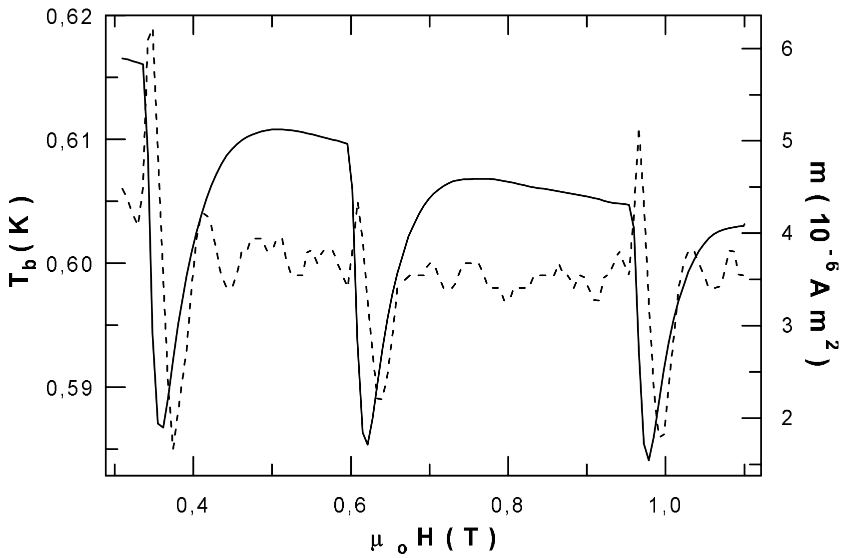

Fig. 2. Full line: magnetic moment during a field sweep experiment. Dashed line: temperature of the electrode (high thermal coupling, 0.69 T/min).

is, for a cylindrical sample:

$$E \approx \frac{1}{2}\mu_0 R\dot{H}_e \qquad (5)$$

where $\dot{H}_e$ is the time derivative of the applied magnetic field. Using the definition of the dynamic relaxation rate as given in [13]:

$$Q = d\mathrm{Ln}(m)/d\mathrm{Ln}(\dot{H}_e) \qquad (6)$$

we obtain:

$$\rho \approx \frac{1}{2}\mu_0 R Q^{-1} J^{-1}\dot{H}_e. \qquad (7)$$

Taking the values: $\mu_0\dot{H}_e \approx 10^{-2}$ Ts^{-1}, $R \approx 10^{-4}$ m, $J \approx 10^8$ Am^{-2} and $Q^{-1} \approx 10$ as will be seen in the rest of this study, we obtain the typical resistivity of a field sweep experiment in our case: $\rho \approx 10^{-13}$ Ωm. The thermal diffusion constant is:

$$D_T = \kappa/C \qquad (8)$$

where κ is the thermal conductivity and C is the thermal capacity which may be estimated from values at 1 K in the literature: $C \approx 10$ $\mathrm{Jm}^{-3}\mathrm{K}^{-1}$ [14] and $\kappa \approx 1$ WK $^{-1}$ m $^{-1}$ [15]. These estimates result in a ratio $D_T/D_E \approx 10^5$ showing that the redistribution of the magnetic flux during an instability is much slower than the heat diffusion through the sample, so that the sample temperature is essentially uniform. A similar conclusion was obtained from the consideration of the parameter values in the high temperature superconductors in [7]. It should be noticed that this should be valid also in the limit $T \to 0$, as one expects a diverging ratio κ/C, limited only by the phonon mean free path cutoff. We now may derive the field increase between two consecutive instabilities. The general criterion for the instability onset is [7]:

$$nK^{-1}\int E\left|\frac{\partial J}{\partial T}\right| dV = 1 \qquad (9)$$

where the integral extends over the sample volume. K is the heat transfer coefficient between the cooling capacitance electrode and the sample. It relates the heat flux through the thermal link to the temperature difference between the sample (assumed uniform in the dynamic approximation) and the electrode:

$$P = K\delta T. \qquad (10)$$

In the rest, $T_b = T - \delta T$ denotes the temperature of the bath. The heat transfer coefficient cannot be known *a priori* and depends on the temperature, the thickness of the thermalizing material as well as on the nature of the boundary surfaces. However, it may be assumed constant at a given temperature. For a cylindrical sample, equation (9) yields:

$$H_j = \left(2J^2K/\pi\mu_0 LRn\dot{H}_e|\partial J/\partial T|\right)^{1/2}. \qquad (11)$$

Here, it was assumed that the current density inside the sample is uniform, *i.e.* the field dependence of the screening current on the scale of the field for full magnetic penetration of the sample (H^*) is negligible (this is clearly not so for larger field variations, as can be seen in Figure 3 showing a strong decrease of the screening current with the applied field).

No instability can occur as soon as $H_j > H^*$, as the total dissipated power is determined in this case only by the product $J\dot{H}_e$ and decreases with the applied field. This is achieved when the screening current density is low enough, which may be realized for the higher magnetic fields, as shown in Figure 1. For the same reason, the recovery magnetization curve after a jump tends to saturate to a field independent value at the higher fields, while the jump occurs on an increasing $m(H)$ curve for the lower ones.

The use of equation (11) requires the knowledge of the screening current density, the non-linearity coefficient $n \approx U_0/kT$ and the temperature derivative $\partial J/\partial T$. The determination of these parameters from the curves in

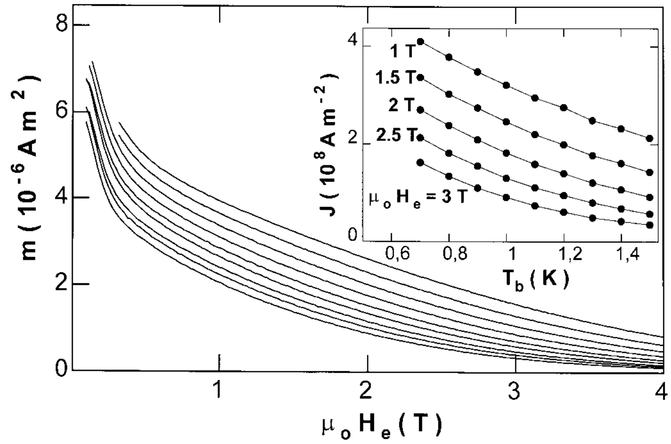

Fig. 3. Screening current density during field sweep experiments (high thermal coupling, 0.6 T/min). From top to bottom: $T_b = 0.7$ to 1.5 K by step of 0.1 K. Insert: same data as a function of the temperature.

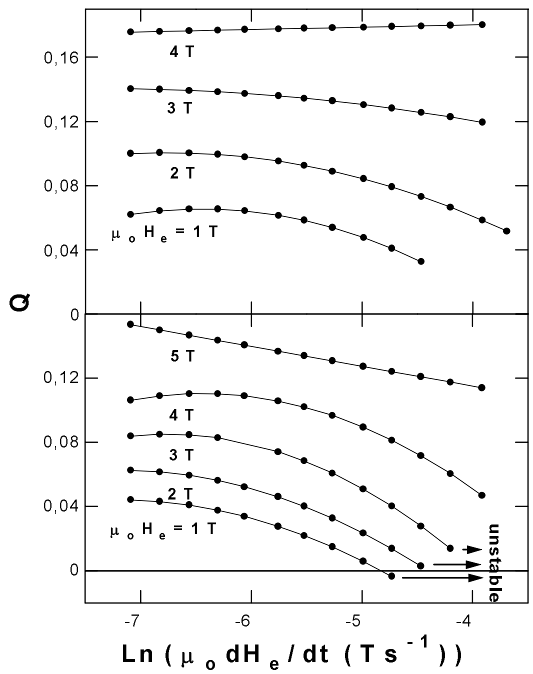

Fig. 4. Dynamic relaxation rate, $Q = d\mathrm{Ln}(m)/d\mathrm{Ln}(dH_e/dt)$ (High thermal coupling). a) $T_b = 1$ K, b) $T_b = 0.6$ K. The arrows indicate the transition to the unstable regime when the sweep rate is increased further.

Figure 1 is not possible, as a full critical state cannot be established as long as instabilities occur. Equation (11) shows that magnetothermal instabilities may be suppressed by an increase of the heat transfer coefficient or the use of thin samples. To this purpose, in a second experiment (referred to as "high thermal coupling"), the thickness of the vacuum grease layer used to thermalize the sample was reduced. We have observed in this case a strongly reduced occurrence of the instabilities. As a result, using the data from above H^*, we could derive the screening current density as well as its temperature derivative for most of the field and sweep rate ranges shown in Figure 1 (Fig. 3). The activation energy and, hence, the non-linearity coefficient may be obtained from the field sweep experiments. As demonstrated in [13], the flux creep equation may be written as:

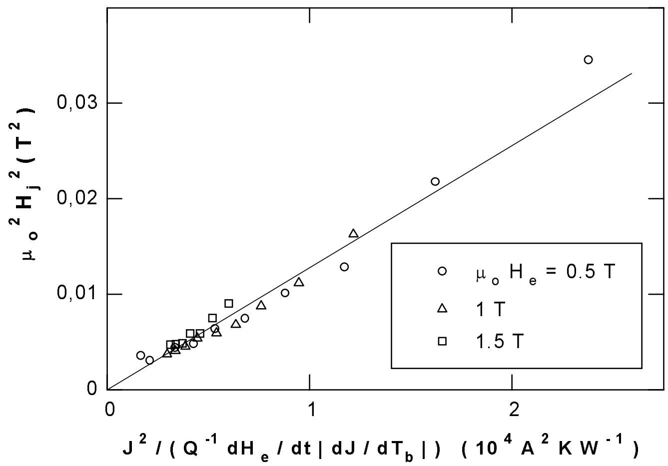

Fig. 5. Plot according to equation (11) for the field increase between two consecutive jumps in Figure 1.

$$U = kT\mathrm{Ln}\left(\frac{2\nu_0 H_e}{R\dot{H}_e - (\Lambda L/\pi\mu_0)(\partial J/\partial t)}\right) \tag{12}$$

where Λ is the self inductance of the cylindrical sample and ν_0 is the velocity of flux lines in the flux flow regime. Using equations (6, 12) where we neglect the second term in the denominator (this can be checked using $\Lambda \approx \mu_0 R\mathrm{Ln}(R/L)$ and the data in Fig. 3), the dynamic relaxation rate may be expressed as:

$$Q \approx -kT\left(\frac{dU}{d\mathrm{Ln}J}\right)^{-1}. \tag{13}$$

In the case of a logarithmic activation energy (Eq. (3)), equation (13) shows that Q should be independent of the sweep rate. As can be seen in Figure 4a, this is a good approximation for the higher magnetic fields, while there is a pronounced downwards curvature for the smaller fields and higher sweep rates. As will be seen below, this does not invalidate the following analysis and we shall assume that $U(J)$ may be described locally by a logarithmic dependence such as the one given by equation (3). According to the discussion above, this is valid provided that $n \approx U_0/kT \approx Q^{-1} \gg 1$, which can be checked readily from the data in Figure 4a. Doing so, we may now apply equation (11), which predicts that H_j^2 should be proportional to the quantity $J^2 Q/\dot{H}_e|\partial J/\partial T|$, the proportionality factor being dependent only upon the dimensions of the sample and the thermal coupling constant. We have determined H_j at three different fields in Figure 1 and plotted the data in this way. The three sets of data are found to coincide roughly on a single straight line, as expected (Fig. 5). The slope of the line may be used to evaluate the thermal coupling constant. This parameter is useful in the determination of the actual temperature of the sample during a field sweep experiment. Equating the cooling power to the dissipated energy during a field sweep, the temperature difference between the coolant and the sample is given, for $H \geq H^*$, by:

$$\delta T = \mu_0 \dot{H}_e K^{-1} m(H_e). \tag{14}$$

From the plot in Figure 5, we estimate $K = 1 \times 10^{-7}$ WK^{-1} for the first experiment. As an example, the temperature difference between the sample and its holder at the last jump for the lowest and highest sweep rates shown in Figure 1 is found respectively $\delta T = 0.05$ K and $\delta T = 0.07$ K. We now show that, although this temperature difference is small as compared to the nominal temperature, it can result in dramatic effects on the apparent flux line dynamics, even in the absence of any instability.

Let us consider the dynamic relaxation rate in the high thermal coupling experimental configuration, at $T = 0.6$ K, for which the higher sweep rates induce instabilities at the lower fields (Fig. 4b). As shown in Figure 4b, Q clearly decreases for increasing sweep rate. In addition, for the lower magnetic fields, Q tends toward zero and, for sweep rates at which it would extrapolate to *negative* values, instabilities similar to the ones described in the low thermal coupling configuration are observed. The decrease towards zero is highly anomalous. As stated above, the exact dependence of the dynamic rate upon the sweep rate is determined by the current dependence of the activation energy. For a given $U(J)$ dependence, equations (12, 13) allow for the determination of Q as a function of $\dot{H}_e$. As an example, here are the expressions for Q, using the main functional dependencies for $U(J)$ encountered in the literature:

$$\begin{aligned}
&U(J) = U_0(1 - J/J_0),\ Q^{-1} = U_0/kT - \mathrm{Ln}(H_e/\tau\dot{H}_e),\\
&U(J) = U_0 \mathrm{Ln}(J_0/J),\ Q^{-1} = U_0/kT\\
&U(J) = \mu^{-1}U_0[(J_0/J)^{\mu} - 1],\ Q^{-1} = U_0/kT + \mu \mathrm{Ln}(H_e/\tau\dot{H}_e).
\end{aligned} \tag{15}$$

In a more general way, Q^{-1} should be finite in the limit $J \to J_0$. Our observation that the instabilities occur as soon as Q reaches zero suggests that the decrease of this quantity is related to the proximity of the unstable regime. Taking into account the sample heating, equation (13) must be replaced by:

$$\begin{aligned}
Q = &-kT(\partial U/\partial \mathrm{Ln}(J))^{-1}\\
&+ (\partial \mathrm{Ln}(J)/\partial T)(\partial T/\partial \mathrm{Ln}\,\dot{H}_e).
\end{aligned} \tag{16}$$

The correction brought in equation (16) by the difference between the temperature of the sample and the one of the electrode is likely at the origin of the downwards curvature observed in Figure 4a. As a consequence, the value derived for n, using equation (13), might be also affected by heating. The observation of the linear behavior in Figure 5 suggests that this had little influence on the determination of the heat transfer coefficient. Finally, using equations (14, 16) and $H^* = H_j = JR$ as the stability criterion, it is easy to show that equation (11) reduces indeed to $Q = 0$.

In this way, the stability criterion presents a striking similarity with the predictions made for flux avalanches. Recently, an increasing number of theoretical, as well as experimental works have pointed out that the relaxation in superconductors may be affected by the presence of avalanches. Bean's critical state is known to be a "self-organized critical" state. It was pointed out by Tang [16] that such a system should present avalanches, which may be viewed as fluctuations around a stationary state. Their typical size should diverge as the system approaches the critical state ($J \to J_0$). As pointed out in [17,18], the avalanches should control the relaxation from the critical state mostly at short times, as the increase of the activation energy away from the critical state prevents the formation of large avalanches. The equivalent for short time in our case is the large sweep rate (for the higher sweep rate, we evaluate the equivalent time for a relaxation experiment to $10^{-4} - 10^{-3}$ s, following [13]). It was shown also in [17,18] that the presence of avalanches should result in a diverging, negative effective activation energy as J approaches J_0. Such a statement, in view of equation (12), is similar to our finding that the dynamic relaxation rate should be zero at the instability threshold. The derivative of the effective activation energy with respect to current is obtained easily from equation (13, 16):

$$\frac{\partial U_{eff}(J)}{\partial J} = \frac{\partial U(J)}{\partial J}\left[1 - \frac{\delta T}{T}\frac{U(J)}{kT}\right]^{-1} \tag{17}$$

where δT is given by equation (14). This shows explicitly that, although $\delta T \ll T$, the internal heating can result in a dramatic change of the critical state dynamics, eventually reaching a catastrophic magneto-thermal instability. This should be true also for relaxation experiments which, although performed in the sub-critical regime, may be strongly affected. Thus, in several aspects, magnetization experiments on samples with a finite thermal coupling to the environment present similarities with the predictions for avalanches. The physics involved is however very different. In particular, in the case studied here, there is an additional feedback mechanism, through the sample temperature, with respect to the avalanche mechanism. Moreover, the consideration upon the ratio of the magnetic and thermal diffusion constants shows that avalanches cannot be due to a temperature rise on the scale of the flux line lattice spacing, following a vortex jump in the flux creep regime. Thus, the consideration of an adiabatic avalanche mechanism, as proposed in [19], is not justified.

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