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Resistive noise and heat effects in the granular oxide superconductor $\text{BaPb}_{0.75}\text{Bi}_{0.25}\text{O}_3$

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Abstract. For weak applied magnetic fields the frequency spectrum of the voltage fluctuations was measured for Josephson ceramic samples of $\text{BaPb}_{0.75}\text{Bi}_{0.25}\text{O}_3$ with $T_c \approx 10$ K. The measurements were carried out in the resistive state. It is shown that the spectral density $S_V(f)$ in the frequency region $20 \text{ Hz} < f < 500 \text{ Hz}$ is described by a function $f^{-\alpha}$, where $\alpha = 1$ for small H and $\alpha = 2-2.5$ for $H \geq 10$ Oe. For currents $I \geq 0.8$ mA through the sample, oscillations with a fundamental frequency $f_1 \leq 100$ Hz and two harmonics f_2 and f_3 were observed. For larger currents these oscillations die out and instead several new oscillations appear. Flicker noise with $\alpha = 1$ is due to the existence of the superconducting glass state. The transition to $\alpha \approx 2$ and the emergence of the oscillations apparently occur owing to heating effects.

1. Introduction

It is well known [1] that, in conventional type-II superconductors, thermally activated creep of the trapped flux is observed. It involves the motion of flux bundles under a magnetic flux gradient. As a consequence, the magnetic flux in a sample decreases or increases (depending on the experimental set-up) logarithmically in time. It is natural that this phenomenon is observed only in the case when Abrikosov vortices penetrate into the bulk of a sample, i.e. for the magnetic field range $H_{c1} < H < H_{c2}$. Here H_{c1} and H_{c2} are the first and second critical fields of the bulk superconductor.

In ceramic superconductors including high- T_c ones the situation is much more interesting. These superconductors involve a disordered porous medium consisting of superconducting grains interconnected by weak links of either the SNS or the SIS type [2–8]. The existence of weak links leads to substantial weakening of the diamagnetic shielding (regime of zero-field cooling) and the Meissner effect (regime of field cooling) [9–12]. Therefore the complete Meissner effect survives only up to a magnetic field [9, 12–14]

$$H_1 = \bar{K}(zI_c\varphi_0/\mu cd^3)^{1/2}. \quad (1)$$

Equation (1) was obtained under the assumption that the real disordered Josephson medium is replaced by an effective averaged Josephson medium. Here \bar{K} is a numerical

constant close to unity, I_c is the typical critical Josephson current of the intergrain junction, $\varphi_0 = hc/2e$ is the flux quantum, h is the Planck constant, e is the elementary charge, c is the velocity of light, d is the typical diameter of a grain or a pore, z is the number of weak links per one grain, $\mu = 1 - f$ is the magnetic permeability and f is the part of the sample filled by the superconducting grains. For $H > H_1$ the 'hypervortices' [10, 12, 14] covering many grains penetrate into ceramics. The field H_1 is several orders of magnitude smaller than the earth's magnetic field. Therefore all measurements in unscreened cryostats are concerned with the peculiar 'mixed' (Abrikosov-type) phase [10].

A further field increase results in the overlapping of the isolated 'hypervortices'. The non-uniform weak-link frustration which takes place for $H \neq 0$ leads to the formation of the non-ergodic superconducting glass (SG) state with a wide continuous spectrum of the relaxation times [10, 11, 13, 15–18]. The main feature of this frustrated system is the existence of many current-carrying diamagnetic states of the porous sample which have the same energy. The peculiar continuous spectrum of the relaxation times corresponds to transitions between local and global energy valleys, i.e. permanent redistribution of Meissner currents between various loops. The SG state is to a large extent analogous to the spin-glass state in magnetics [19]. However, contrary to the latter, the degree of frustration in the SG state depends on H and therefore changes itself during the relaxation process. The field of the SG-phase formation is usually thought [10, 11, 13] to be $H_{cg} = \varphi_0/G \ll H_{c1}$, where $G = d^2$ is a typical pore cross section. However, the SG-like properties can be really observed in the non-homogeneous medium even for $H < H_{cg}$. On the other hand, the oscillation phenomena connected with the existence of grains of typical size d appear only when H reaches H_{cg} from below and vanish for $H \gg H_{cg}$ [17]. The oscillations with a period proportional to H/H_{cg} of the critical current I_c were observed initially for Josephson-type samples of $\text{BaPb}_{1-x}\text{Bi}_x\text{O}_3$ (BPB) [3, 5]. In this case these oscillations were also shown to die out for large H . The oscillation damping of the various quantities for $H \gg H_{cg}$ because of their spatial averaging was predicted in the early work of Rosenblatt and co-workers [9, 20]. If the frustration is uniform, damping should be absent [21].

Thus, non-ergodic behaviour can be observed in ceramics over two magnetic field ranges:

- (i) $H_{cg} \leq H \leq H_{c1}$;
- (ii) $H_{c1} \leq H \leq H_{c2}$.

Our experiments which are discussed below concern mainly range (i) for the SG state. Note that above we have suggested implicitly the validity of the Josephson model with a Hamiltonian [10, 15–17, 20]:

$$\mathcal{H} = - \sum_{ij} J_{ij} \cos(\varphi_i - \varphi_j - A_{ij}) \quad (2)$$

$$A_{ij} = \frac{2\pi}{\varphi_0} \int_i^j \mathbf{A} \cdot d\mathbf{l}. \quad (3)$$

Here φ_i is the order parameter phase in the i th grain; \mathbf{A} is the vector potential; the quantities J_{ij} depend on temperature T and the weak link type (SNS or SIS). Nevertheless, experiment has shown [22] that at least the relaxation properties of the porous medium are not changed if a sample is annealed, so that instead of the Josephson model (2) the percolation network model [23] becomes more appropriate. According to our

knowledge, the macroscopic manifestations of the irreversible dynamics and the equilibrium noise of the superconducting granular system do not depend on the choice of the model.

The non-equilibrium SG state studies in various measurements of the magnetization time dependences is characterized, as was stated above, by the extremely wide range of the relaxation times τ . The real duration of the measurements makes the relaxation time spectrum effectively infinite. Therefore, the time dependence of the thermoremanent magnetization M_{TRM} in different ceramic superconductors for fields $H_{cg} \leq H \leq H_{c1}$ obeys the law [5, 22, 24, 25]

$$M_{TRM}(t) = M_{TRM}^0 - S^R \log(t/\tau_0). \quad (4)$$

Here τ_0 is the initial transient time interval; $S^R = S^R(T, M_{TRM}^0)$ is the magnetic viscosity. In this case for constant T the universal dependence $S^R \approx (M_{TRM}^0)^3$ is observed for various substances [26]. We note that memory effects which alter the dependence (4) and are so representative for the glass states [19] have also been observed for the high- T_c oxide $YBa_2Cu_3O_{7-\delta}$ [27] but after the field jump $\Delta H = 1 \text{ kOe} > H_{c1}$ and for a single-crystal specimen. For the SG field range, memory effects have not as yet been detected.

As was shown by the theory [19, 28] developed for spin glasses, the wide relaxation time spectrum should lead not only to the relaxation law (4) but also to the appearance of excess equilibrium magnetic $1/f$ (flicker) noise superposed on the conventional thermal noise. Here f is a frequency. The spectral density $S_M(f)$ of the flicker noise and the relaxation law $M_{TRM}(t)$ (after the constant field H is switched off) turn out to be connected by a certain relationship. In fact, according to the Wiener-Khinchin theorem, we have

$$\begin{aligned} S_M(f) &= 2 \int_{-\infty}^{\infty} dt \exp(i\omega t) \langle M(t)M(0) \rangle = 4 \int_0^{\infty} dt \cos(\omega, t) \langle M(t)M(0) \rangle \\ &= -\frac{4}{\omega} \int_0^{\infty} dt \sin(\omega t) \frac{d}{dt} \langle M(t)M(0) \rangle. \end{aligned} \quad (5)$$

Here $M(t)$ is an equilibrium fluctuating magnetization at the moment t ; $\omega = 2\pi f$ is a circular frequency; $\langle \dots \rangle$ means the ensemble averaging. On integrating equation (5) by parts the first term vanished owing to the absence of the fluctuation correlations for infinitely distant times. On the other hand, the fluctuation-dissipation theorem leads to the following equation [29]:

$$\langle M(t)M(0) \rangle = K(t)T. \quad (6)$$

Here the Boltzmann constant k_B is considered to be unity and the relaxation function $K(t)$ is determined by the response of the mean magnetization value to the magnetic field which is equal to H for $t \leq 0$ and to zero for $t > 0$ (such a set-up corresponds to the thermoremanent magnetization):

$$K(t) = M_{TRM}(t)/H. \quad (7)$$

From (5-7) taking into account the relaxation law (4) we obtain

$$S_M(f) = -\frac{2T}{\pi f H} \int_0^{\infty} dt \sin(2\pi ft) \frac{d}{dt} [M_{TRM}(t)] = \frac{2TS^R}{\pi f H} \int_0^{\infty} \frac{dt}{t} \sin(2\pi ft). \quad (8)$$

For real systems the relaxation time spectrum is bounded both above and below. Then in the frequency range $\tau_{\max}^{-1} \ll 2\pi f \ll \tau_{\min}^{-1}$ (where τ_{\max} and τ_{\min} are the maximum and

minimum relaxation times, respectively) for systems that obey equation (4) the flicker noise spectral density is determined by the relationship [28]

$$S_M(f) = S^R T/Hf. \quad (9)$$

The experiment confirmed the existence of such noise in spin glasses [19]. One should expect the same magnetic noise to be observed in the superconducting glasses—ceramic superconductors.

For low frequencies $f \geq 1-10^3$ Hz and external magnetic fields $H \geq 0.5$ Oe the noise spectrum was measured in different high- T_c superconductors (ceramics and single crystals) by a number of groups (see, e.g., [30, 31]) with the help of SQUID magnetometers. The flicker noise connected with the flux redistribution between Josephson network loops was in fact observed in these experiments. Its magnitude and spectral characteristics depend on H substantially.

Unfortunately, in the high- T_c ceramics the situation is influenced by existence of twins in separate grains [6-8, 32]. That is why the relative role of different size structure elements in these Josephson media is unclear. It makes the interpretation difficult in the framework of the SG model. The Josephson ceramic specimens of BPB with $T_c = 10$ K and $H_{c1} \approx 12$ Oe [4] for $x = 0.25$ are free from this fault. They are investigated in this work. In contrast with [30, 31] we use, however, the resistive method of measurement. Such an experimental set-up is based on the natural hypothesis that magnetic flux fluctuations influence the resistance of the current-carrying infinite cluster in the percolation grain system. Then the magnetic noise spectral density $S_M(f)$ is proportional to the spectral density $S_V(f)$ of the voltage fluctuations.

Note that the four-probe method is sensitive to a magnetic flux redistribution in the bulk of the sample whereas SQUID magnetometers respond only to the overall sample's magnetic flux variation and may record the superconducting screen noise. Then the interpretation becomes ambiguous [33]. Furthermore, we should emphasize the importance of the resistive noise investigations from the practical point of view, because the operation of bolometers or superconducting switches is accompanied by the recovery of the resistive state.

2. Experimental procedure and results

The samples in the form of parallelepipeds $1.5 \text{ mm} \times 2 \text{ mm} \times 12 \text{ mm}$ were incorporated into the four-probe circuit. A finite resistance was obtained in two ways:

- (i) switching from the superconducting branch to the single-particle branch for forward current-voltage (I - V) characteristics;
- (ii) operating with reverse I - V characteristics.

The noise was analysed with a SK4-72 spectrum analyser in the frequency range $20 \text{ Hz} < f < 500 \text{ Hz}$. The current across the sample was determined by the battery current source with the help of an RC filter.

The waiting time τ_0 for realization of the equilibrium noise spectrum was 3-4 min. We see that this quantity is well above $1/f_{\min} = 0.05$ s. The margin seems to be sufficient because, in similar systems (spin glasses), noise equilibrium was achieved for $\tau_0 \geq 10^3/f_{\min}$ [19].

Note that, if one bears in mind our usage of the resistive method, then the ceramics BPB possesses two advantages compared with the high- T_c superconductors:

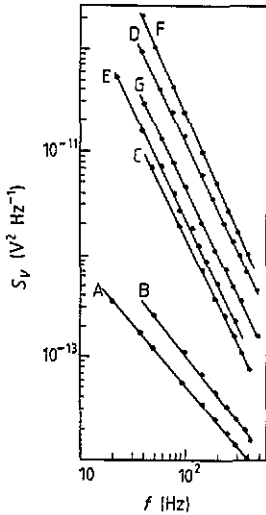


Figure 1. The frequency f dependence of the resistive noise spectral density S_V for forward I - V characteristics: curves A-C correspond to cooling in the field $H = H_c$ where the operating currents are $I_1 = 5$ mA, $I_2 = 6$ mA and $I_3 = 7$ mA, respectively; curves D and F correspond to cooling in the fields $H = 5$ Oe and 10 Oe, respectively; curves E and G correspond to when the fields $H = 5$ Oe and 10 Oe are switched off; $I = I_1 = 5$ mA for curves D-G ($T = 4.2$ K).

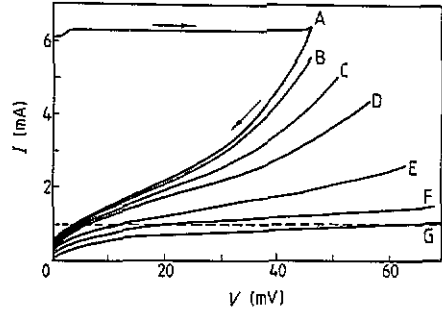


Figure 2. I - V characteristics in various fields H : curve A, 0 Oe; curve B, 0.5 Oe; curve C, 1 Oe; curve D, 2 Oe; curve E, 5 Oe; curve F, 10 Oe; curve G, 50 Oe; ---, operating points with $I = 1$ mA on the I - V characteristics.

- (i) a relatively large electrical resistivity;
- (ii) tunnel-like (not smooth) switching between branches of the I - V characteristics.

In fact, the resistive noise measurements for $YBa_2Cu_3O_x$ were carried out only in the neighbourhood of T_c [34]. Moreover, it seems to us that the figures in [34] are in contrast with the conclusion of [34] that noise spectra above and below T_c are different.

In figure 1 the frequency dependences of $\log S_V$ are shown. They were obtained by measurements of the forward I - V characteristics. Curves A and B represent the dependence $S_V \sim 1/f^{1.5}$ obtained after cooling the sample in the earth's magnetic field with the vertical component $H_c \approx 0.5$ Oe. This noise vanishes above T_c , i.e. it is directly linked to the occurrence of the superconducting state. The dependences of curves A and B correspond to the operating points of the I - V characteristics with $I_1 = 5$ mA and $I_2 = 6$ mA. When the operating current is equal to $I_3 \approx 7$ mA the dependence of $\log S_V$ on $\log f$ has a different slope than for smaller currents, so that the corresponding curve C describes the law $S_V \sim f^{-2.5}$.

The measurements for $I_1 = 5$ mA were also carried out in the applied magnetic field $H = 5$ Oe oriented along the sample. In this case the cooling process was made in the magnetic field (field-cooling regime). As can be seen from figure 1 (curve D), the dependence $S_V \sim 1/f^{2.5}$ is then realized. After the magnetic field has been switched off, the I - V characteristics do not recover owing to the partial magnetic flux trapping. However, the frequency dependence of the spectral density is not changed (curve E). Similar behaviour was obtained for the magnetic field $H = 10$ Oe after cooling (curve

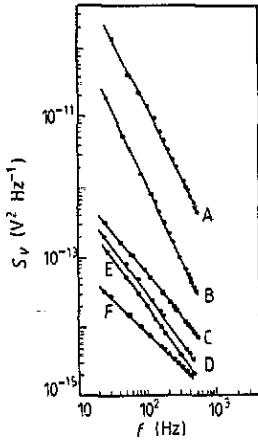


Figure 3. The same as in figure 1 for the reverse I - V characteristics for various H : curve A, 16.6 Oe; curve B, 10 Oe; curve C, 0 Oe; curve D, 5 Oe; curve E, 2.3 Oe; curve F, 0.6 Oe.

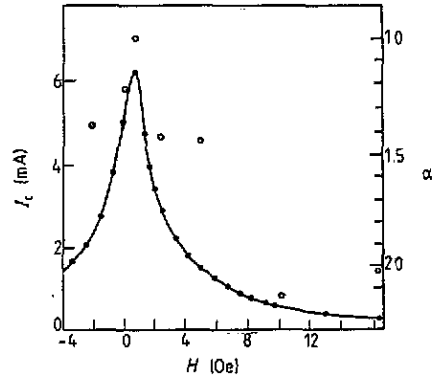


Figure 4. The dependence on H of the exponent α for $S_V(f) \sim f^{-\alpha}$ (O) and of the critical current I_c (—).

F) and when the field is switched off (curve G). We note that the noise amplitude is larger for larger operating current, magnetic field or trapped magnetic flux. It agrees with the non-linear behaviour of the magnetic viscosity $S^R(M_{\text{TRM}}^0)$ which was discovered by us earlier [22, 26]. Indeed, M_{TRM}^0 in BPB is proportional to H for $H \leq 20$ Oe [22]. Then from (9) and taking into account the dependence $S^R \sim H^3$ [26] we obtain

$$S_M(f) \sim H^2 T / f. \quad (10)$$

In the new series of measurements the resistive noise was studied for the reverse I - V characteristics. Another sample of BPB with $x = 0.25$ was studied. The I - V characteristics for different values of the applied field H are presented in figure 2. The chosen operating current value $I = 1$ mA is shown by the broken curve. For $H > 0$ the forward I - V characteristics are not shown.

The measurements showed that in this case the quantity S_V obtained after cooling the sample in the earth's magnetic field depends on the frequency according to the considered above power law: $S_V \sim f^{-\alpha}$ (figure 3). The exponent α depends on the applied field H and the operating current I non-monotonically. This noise disappears above T_c in the same way as the noise described in figure 1.

In figure 4 the values of α for different H and $I = 1$ mA are shown by open circles. For comparison the dependence on H of the sample's critical Josephson current is shown here by full circles and the full curve. From figure 4, one can see that the dependences $\alpha(H)$ and $I_c(H)$ correlate well. Note that the asymptotic value of $I_c(H)$ for large H (see also [4]) corresponds to the so-called percolation current as distinguished from the coherent current [9]. The latter dominates for small H .

In both series of measurements the $1/f$ noise is observed in the region of maximum Josephson currents and small magnetic fields, where the SG state should be realized [15–18]. In this case, one has to bear in mind that for the given experimental conditions the effective field always exceeds H_{c0} in any nominal external magnetic field. Resistance fluctuations with the $1/f$ spectrum may be a consequence not only of the magnetic flux

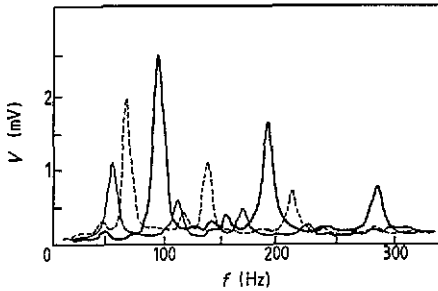


Figure 5. The frequency spectra of the voltage oscillations (without the background contribution) for operating currents I of 1.4 mA (—), 1.2 mA (---) and 1.1 mA (-·-·-).

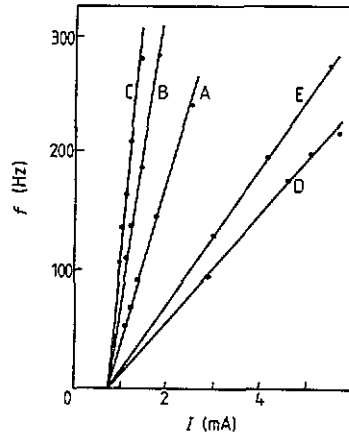


Figure 6. The dependences on I of the fundamental frequency f_1 (curve A), its harmonics f_2 (curve B) and f_3 (curve C) and the frequencies f_4 and f_5 of new oscillations (curves D and E).

redistribution between Josephson medium cells [24], but also of the random formation and destruction of the fluctuating current circuits. Such fluctuations are more probable in those sites where T_c is lowered owing to local variations in the bismuth content. Similar magnetic fluctuations were investigated near the superconducting transition in samples of $YBa_2Cu_3O_{7-x}$ screened from the external magnetic field H [35].

The results of the observations described above show that an increase in the effective field modulus leads to an increase in the exponent. Since all the applied field values except $H = 16.6$ Oe are less than $H_{c1} = 12$ Oe for the single crystal of BPB [36], the flux creep phenomenon inside the grains should not influence the results. Therefore, the transition from the $S_V \sim 1/f$ law to the $S_V \sim 1/f^2$ dependence when H increases seems to stem from either of two mechanisms:

- (i) the occurrence of the oriented motion of the magnetic flux quanta affected by the Lorentz force (the flux flow regime [1]);
- (ii) the destruction of the macroscopic superconducting circuit number, i.e. the disappearance of the SG state.

In the latter case the exhaustion of the superconducting current network may lead to manifestation of thermal effects due to the Joule heating. Then the spectrum $S_V(f) \sim 1/f^2$ will correspond to temperature fluctuations [37]. This interpretation agrees well with the increase in α when the operating current I increases (see figure 1). The observed increase can be explained naturally by the heating effects.

For large enough I the spectra $S_V(f)$ in the second series of measurements incorporate not only the background dependence $f^{-\alpha}$ but also contributions from some low-frequency oscillations. The dependences on f of the fluctuation voltage amplitude V for various values of I are shown in figure 5. Three harmonics can be easily distinguished. Their amplitudes increase when I increases. Further increase in the current causes these oscillations to vanish and other oscillations (f_4 and f_5) to appear. The frequencies f_i of the oscillations increase for larger I .

The dependences $f_i(I)$ are shown in figure 6. All of them are almost linear. The oscillations f_1 , f_2 and f_3 do not exist when the current I is less than the threshold value I_{th} , which is about 0.8 mA for the sample considered. The quantity I_{th} seems to coincide with the re-entrant supercurrent. This indicates that the I - V characteristics is to a great extent determined by the heating effects (as was stated above).

The low frequency $f_1 = 100$ Hz of the observed oscillations and the presented speculations about the thermal effect role in the electrophysical properties of the ceramics BPB lead to the assumption that these oscillations are of a thermal nature and involve relaxation self-sustained oscillations. They are analogous to the oscillations in single Josephson junctions [38, 39]. The main argument is the existence of the threshold current I_{th} . The non-linear self-sustained oscillations in Josephson junctions with heating are quite similar [37, 39] to the relaxation oscillations in the junctions with a capacity-driven hysteresis which work under conditions of the given voltage [38, 39]. In our case such a regime is apparently realized in single Josephson junctions in the bulk of the sample, the whole sample being the voltage source. The object complexity makes it difficult to compare the linear dependences $f_i(I)$ with the dependences [38] of the oscillation period on the mean voltage \bar{V} at the junction and the current \bar{I} across the junction. One should bear in mind that for high currents the heating effects begin to play the main role and the earlier assumed proportionality between $S_M(f)$ and $S_V(f)$ no longer occurs.

3. Summary

To summarize, we emphasize that our experiments demonstrate both the glass-like behaviour of the Josephson medium—the ceramics BPB—and the importance of the thermal effects for large currents across the sample. The necessity to take account of the heating phenomena in current-carrying ceramics was initially stressed in [3].

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