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Mechanism of Formation of a Negative Magnetoresistance Region in Granular High-Temperature Superconductors

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Abstract—The field dependences of the magnetoresistance of $Bi_{1.8}Pb_{0.3}Sr_{1.9}Ca_2Cu_3O_x$ samples with different densities, which have a foam microstructure and exhibit different diamagnetic responses, were studied at 77.4 K to identify the mechanism responsible for the formation of a negative magnetoresistance region in granular high-temperature superconductors. A region with negative magnetoresistance was found to exist in samples with magnetizations highest in absolute magnitude. This behavior finds a reasonable interpretation as due to the effect exerted by dipole moments of high-temperature superconductor grains on the effective intergranular field. The strength of this effective field has been estimated.

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1. INTRODUCTION

It is known that the magnetic field dependence of the electrical resistance $R(H)$ of granular high-temperature superconductors (HTSCs) features in some cases a region of negative magnetoresistance [1–6]. Grain boundaries in granular HTSCs play the part of Josephson weak links. Therefore, their critical parameters (critical current density, first and second critical fields H_{C1J}, H_{C2J}) are lower than those for the HTSC grains. In most experiments, transport current does not initiate dissipation in the superconducting grains [3–9]. Although it was shown [2, 3] that the negative magnetoresistance region in the $R(H)$ dependence originates from magnetic flux redistribution at grain boundaries in the vicinity of the first critical intragrain field H_{C1G}, nevertheless, the conditions favoring the onset of this feature still remain largely unknown. The dependence $R(H)$ reflects essentially the resistive response of the system of grain boundaries to the effective field in the intergranular medium B_{eff}, which is a superposition of the external field (for $H \geq H_{C1J}$) and of the field induced by the HTSC grain dipole moments [3, 7, 9–1]. To estimate the magnetic field induction in the intergranular medium B_{eff} and understand the mechanism responsible for the occurrence of the negative magnetoresistance region in the dependence $R(H)$, we believed it reasonable to investigate polycrystalline HTSC samples of the same composition but with a modified microstructure and different values of the diamagnetic response.

We report here on a study of the magnetoresistive properties of bismuth HTSCs of different densities which differ in diamagnetic response, and on a first observation of a correlation between the existence of a negative magnetoresistance region with the value of sample magnetization. The objects studied belong to the class of foam HTSCs [12] with a characteristic flake-like grain structure which have micropores [13]. An earlier study dealt with the magnetic properties [14], *I–V* characteristics in zero external field [15] and the resistive transition in a magnetic field [16] in these samples. Thus, the present investigation is also actually a logical continuation of our earlier research.

2. EXPERIMENTAL TECHNIQUE

The samples of the $Bi_{1.8}Pb_{0.3}Sr_{1.9}Ca_2Cu_3O_x$ microfoam intended for experiments were prepared by the technology described in [13, 15, 16]. A study of the microstructure of these samples, which is also covered in the above references, showed the samples to be made up of randomly arranged platelet crystals with linear dimensions of 20–30 μm and 1–2 μm thick. The samples studied had densities of 2.26 and 1.55 g/cm^3 (38 and 26% of the theoretical density, respectively). We shall refer subsequently to these samples as foam 1 (2.26 g/cm^3) and foam 2 (1.55 g/cm^3). We used as reference a dense sample of the same composition (labeled in what follows as poly-Bi). It was prepared of the microfoam by milling, pressing and subsequent sintering at the same temperature as used with the foam (825°C) for 5 h. The density of the poly-Bi sample was found to be 5.28 g/cm^3 (90% of theoretical value). Scanning electron microscopy of this sample revealed a distinct granular microstructure, with

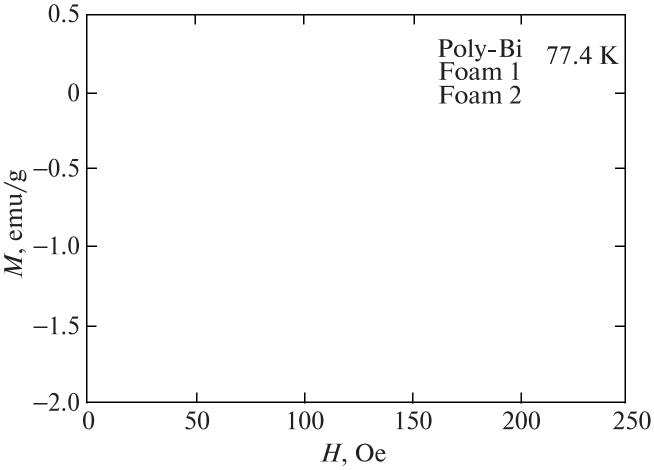

Fig. 1. Magnetization hysteresis in the samples measured at $T = 77.4$ K.

the linear grain dimensions reduced compared with those for the foam (5–10 μm), which was paralleled by an increase in the fraction of small crystallites (up to ~5 μm). X-ray diffraction measurements revealed the dominant phase to be 2-2-2-3, whereas the fraction of the 2-2-2-1 low-temperature phase was less than 5%. Magnetic measurements showed the transition temperature for all the three samples to be 108 K, with the temperature of transition to the $R = 0$ state, as derived from resistive measurements, being 106 K.

Measurement of the magnetoresistance $R(H) = U(H)/I$, where U is the voltage drop across the sample, and I the transport current, $\mathbf{I} \perp \mathbf{H}$, were performed by the standard four-probe technique. A sample measured typically 1 × 1.5 × 8 mm. The electric contacts were prepared using the epoxy resin-based Epo-Tek H20E cement. The sample to be studied was placed in liquid nitrogen. The critical current density at $T = 77.4$ K, as derived from the 1 μV/cm criterion, was ≈0.5, 35, and 80 A/cm^2 for the foam 2, foam 1 and poly-Bi samples, accordingly (the transport critical current I_C through the sample was, respectively, 15, 600, and 410 mA). The electrical resistivity at 120 K (above T_C) was about the same for all the samples, ~1.6 Ω cm. The data on $R(H)$ are given in milliohms. The magnetoresistance at 77.4 K measured in fields above 1 kOe and transport currents of 500–700 mA are about 10% of R(120 K). The samples were cooled in zero field conditions (with no special measures on screening from the Earth's magnetic field taken). After each measurement cycle including increase of the field to the maximum level, $H_\uparrow \longrightarrow H_{max}$, followed by the decrease to $H_\downarrow \longrightarrow 0$ ($H_\uparrow$ and $H_\downarrow$ identify the increasing and decreasing field), the sample temperature was raised above T_C.

The magnetization studies were performed on a vibrating sample magnetometer [17], on the same samples that were used in $R(H)$ measurements, of which the central part (which corresponded to the operating section between the potential contacts) was cut out.[1]

3. RESULTS AND DISCUSSION

Figure 1 shows magnetization hysteresis loops $M(H)$ of the samples under study measured at $T = 77.4$ K. We readily see that porous samples exhibit higher values of the magnetization than the reference poly-Bi sample. Such a behavior was observed earlier [13, 14] and can be attributed to at least two factors. First, the grains in microfoam have fairly large linear dimensions, which gives rise to an increase of the diamagnetic signal due to the intragrain current. Second, in a dense ceramic the grains located in the bulk can be partially screened, a situation that cannot exist in a porous sample. The mechanisms that underpin enhancement of the diamagnetic response in our microfoam samples will be analyzed in a separate publication; the emphasis here will be placed instead on the interrelation between the hysteretic behavior of the magnetization and of the magnetoresistance $R(H)$.

Figure 2 plots the dependences $R(H)$ for the foam 2 sample measured at different transport currents I, in fields of up to 250 Oe and $T = 77.4$ K. These dependences reveal interesting features. First, a clearly pronounced hysteresis with increasing and decreasing external field H. Second, the dependence $R(H)$ obtained in the up run shows a distinct maximum followed by the negative magnetoresistance region. As the field is increased still more, the dependence $R(H)$, on passing through a minimum, starts to grow weakly (Fig. 3). We note also that the dependences $R(H)$ in Figs. 2 and 3 start from the point $R(H = 0) \neq 0$. The reason for this lies in that the measuring current I for the sample under study is larger than the critical current $I_C(H = 0)$ ($I_C \approx 15$ mA).

The dependences $R(H)$ for the foam 1 sample are plotted in Fig. 4. While these dependences likewise exhibit hysteresis, we do not see in them a negative magnetoresistance region. At low enough transport currents ($I = 30$–40 mA) one can discern a very small decrease in resistance occurring with increasing external field, while at large I and at $H = 150$–250 Oe the resistance practically does not vary (in high fields, R also grows, similar to the behavior in Fig. 3).

The hysteresis dependences $R(H)$ for the poly-Bi dense sample do not have a negative magnetoresistance region, which is seen from Fig. 5. The data displayed in Fig. 5 were obtained both for $I < I_C$ ($H = 0$) and for $I > I_C$ ($H = 0$). Our measurements show that,

[1] The authors did not observe any substantial effect (above 10%) of the sample shape (cylinder, plate), for **H** perpendicular to the cylinder axis or to the plate plane, on either the shape or maximum value of the dependence $M(H)$; for the samples studied, the effect of demagnetizing factor plays an insignificant part in estimation of the effective field in intergranular medium.

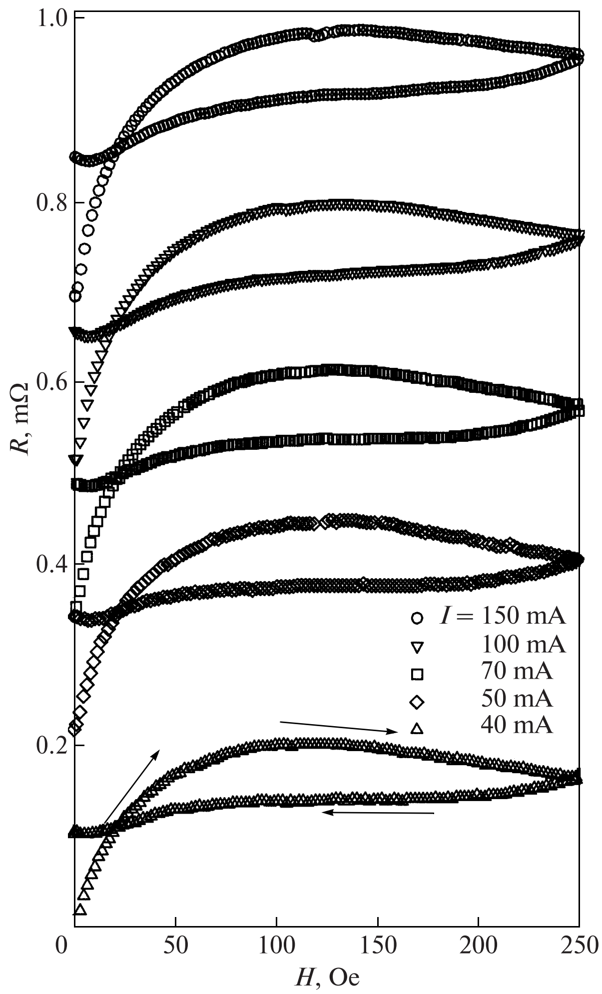

Fig. 2. Hysteresis in the magnetoresistance $R(H)$ of foam 2 sample measured at $T = 77.4$ K, $H_{max} = 250$ Oe, and different transport currents I. The arrows specify the direction of variation in the external field H.

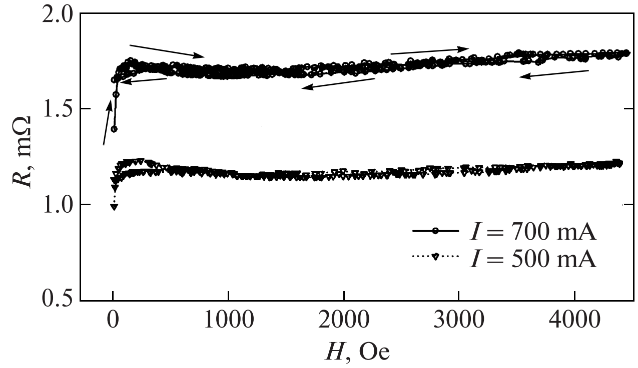

Fig. 3. $R(H)$ hysteresis of foam 2 sample measured at $T = 77.4$ K, $H_{max} \approx 4.4$ kOe, and different transport currents I. The arrows specify the direction of variation in the external field H.

within the field interval 0.25–1.50 kOe, these dependences continue to grow.

Our data on the dependences $R(H)$ and the $M(H)$ (Fig. 1) suggest convincingly that the negative magnetoresistance region is observed in the sample with the largest diamagnetic response. The external magnetic field at which a local magnetoresistance maximum is observed, ~130 kOe, correlates with the field of the minimum in the direct $M(H)$ run (~140 Oe). To understand this result, consider qualitatively the field pattern in the intergranular medium.

Let two adjacent grains possess dipole moments $\mathbf{M}_G(H)$, and the transport current in intergranular space be directed perpendicular to the external field. If the external field increases, the $\mathbf{M}_G$ vectors are oriented against it [9, 11]. Now the field induced by the grain dipole moments in intergranular space, $\mathbf{B}_{ind}$, will amplify the external field, $\mathbf{B}_{ind} \parallel \mathbf{H}$ (the magnetic induction lines from $\mathbf{M}_G$ close across the grain boundary [9, 11]). The effective field in the intergranular medium can be written as $\mathbf{B}_{eff} = \mathbf{H} + \mathbf{B}_{ind}$; hence,

$$\mathbf{B}_{eff} = \mathbf{H} + 4\pi\mathbf{M}_G\alpha, \quad (1)$$

where α is a coefficient determined by the demagnetizing factors of the grains and the dimensions of intergranular spaces. The values of $\mathbf{M}_G$ and α are unknown. The effective field can, however, be analyzed using experimental data on the magnetization $M(H)$ of the samples. In this case, Eq. (1) can be recast in the form

$$\mathbf{B}_{eff} = \mathbf{H} + 4\pi\mathbf{M}(H)\alpha(H), \quad (2)$$

where M is the sample magnetization (in Gauss units), and α is, in a general case, a function of H. Equation (2) explains readily the hysteretic behavior of magnetoresistance, because, as seen from Fig. 1, $|M(H\uparrow)| > |M(H\downarrow)|$, whence $\mathbf{B}_{eff}(H\downarrow) > \mathbf{B}_{eff}(H\downarrow)$; therefore, $R(H\uparrow) > R(H\downarrow)$. In the low-$H\downarrow$ field region, magnetoresistance passes through a minimum or zero value when the induced field exceeds the external field, $|\mathbf{B}_{ind}| > |\mathbf{H}|$. Accordingly, a maximum in the diamagnetic response in the dependence $M(H\uparrow)$ can appear in the dependence $B_{eff}(H\uparrow)$ as a local minimum; hence, magnetoresistance will also pass through a local minimum and, subsequently, a negative magnetoresistance region.

Figure 6 displays graphs relating the effective field in intergranular medium with external field which were plotted for our samples using Eq. (2), with due allowance for the experimental dependences $M(H)$ (emu/g units were reduced to Gauss units using the theoretical density of Bi2223, because pores do not contribute to diamagnetic response). We assumed α = const as a first approximation. It was found that the local maximum in the dependence $B_{eff}(H\uparrow)$ for the foam 2 sample appears for $\alpha > 6$. The dependences $B_{eff}(H\uparrow)$ were constructed using the same value $\alpha = 6.5$

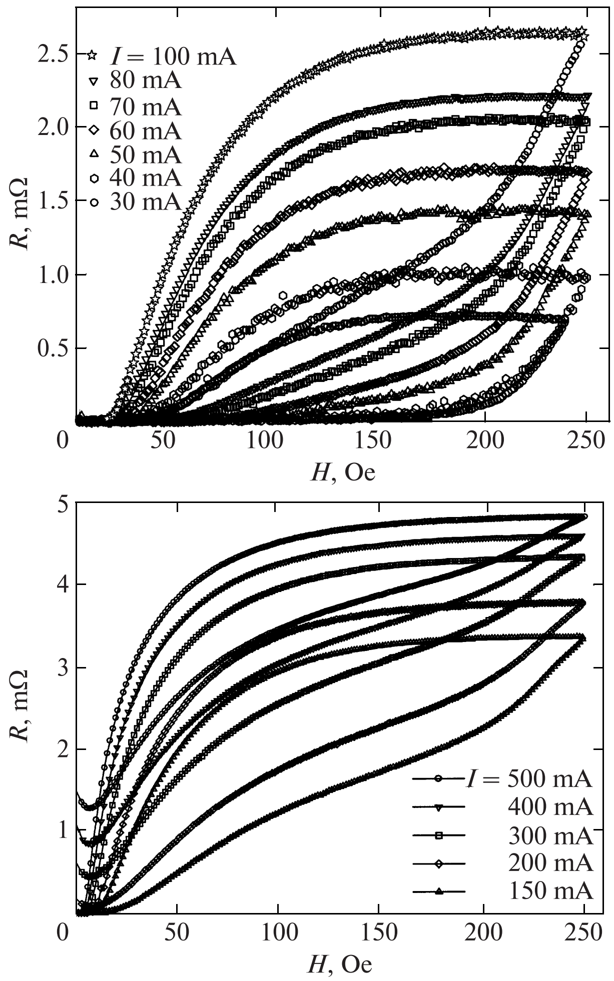

Fig. 4. $R(H)$ hysteresis of foam 1 sample measured at $T = 77.4$ K, $H_{max} = 250$ Oe, and different transport currents I.

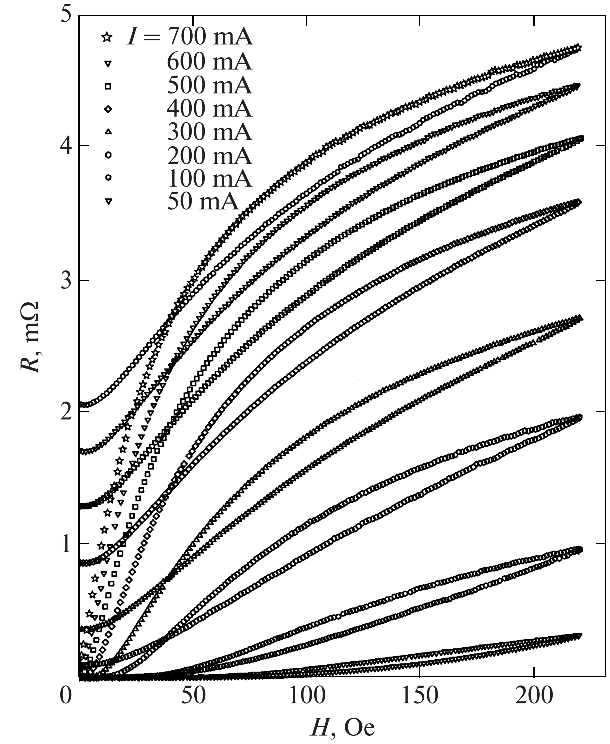

Fig. 5. $R(H)$ hysteresis of poly-Bi sample measured at $T = 77.4$ K, $H_{max} = 250$ Oe, and different transport currents I.

for all the samples.[2] As seen from Fig. 5, for this value of α, the dependences $B_{eff}(H)$ correlate satisfactorily with such features in the dependences $R(H)$ as the appearance of a local maximum for the foam 2 sample, the weakly pronounced local maximum for foam 1, and the absence of this maximum for the poly-Bi sample. The absolute magnitude of the diamagnetic response of HTSC grains provides a dominant contribution to the effective field in intergranular medium and, ultimately, affects the magnetoresistance and the existence itself of the negative magnetoresistance region. This is a crucial point supporting the interpretation of the formation of a negative magnetoresistance region in the dependence $R(H)$ for granular HTSCs. Magnetoresistance can pass through a local maximum in a field close to the minimum in the dependence $M(H_{\uparrow})$ rather than near the field at which vortices start to penetrate into the grains, H_{C1G}, as pointed out in [2, 3]. The H_{C1G} field estimated from data in Fig. 1 as the field at which the $M(H_{\uparrow})$ relation starts to deviate from linear course is ~30–40 Oe for

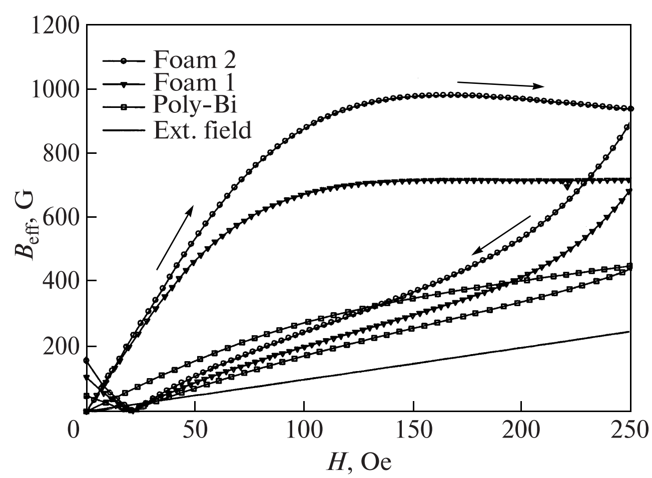

Fig. 6. Effective field in the intergranular medium $\mathbf{B}_{eff} = |\mathbf{H} + 4\pi\mathbf{M}(H)\alpha|$ calculated from the experimental dependences $M(H)$ (Fig. 1) and plotted vs. external field for the samples studied. The arrows specify the direction in the variation of external field H.

[2] For the foam 1 and foam 2 samples this is valid because it is the cleavage planes of the superconducting grains that serve as boundaries in the microfoams, and they are identical for these samples; in the poly-Bi sample, the grain boundary geometry is different, so that α possibly assumes a different value. This does in no way, however, affect the conclusions drawn in this paper.

the samples studied (Fig. 1). At the same time, the values of the external field $H_{\uparrow}$ at which the dependences $B_{eff}(H)$ and $R(H)$ have maxima are close in magnitude (Figs. 2, 4, 5).

Interestingly, the effective field in intergranular medium of a granular HTSC is substantially higher than both the external field and the magnetization of the sample itself (in Gauss units) (Fig. 5). In our opinion, this is a manifestation of magnetic flux compression in intergranular medium of a granular HTSC, which was assumed to exist by some authors [3, 18] but thus far has not been verified experimentally.

Thus, the study of the magnetoresistive properties of $Bi_{1.8}Pb_{0.3}Sr_{1.9}Ca_2Cu_3O_x$ microfoams of various densities has revealed a correlation between the existence of a negative magnetoresistance region in the dependences $R(H)$ and the magnitude of the diamagnetic response. The formation of a negative magnetoresistance region is mediated by the effect of the field induced by the dipole moments of HTSC grains on the effective field at grain boundaries, and it occurs typically in samples with a large diamagnetic response. The compression of the magnetic flux in intergranular medium has been estimated.

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