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MAGNETIC AND MAGNETIC FLUX EXCLUSION PROPERTIES OF SUPERCONDUCTIVE FINE FERRIMAGNETIC POWDER COMPOSITES[1]

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The influence of internal magnetic field on the superconductive properties of composites consisting of a mixture of a superconducting powder such as YBa_2Cu_3-O_{7-x} (particle size is $5 - 15\mu m$) and fine ferrimagnetic particles of Fe_3O_4 ($10nm$ in diameter) was studied as a function of concentration of magnetic particles and their magnetic state. The experimental data were discussed in the framework of the theory where a distribution of internal magnetic fields was taken into account.

1. Introduction

In papers [1,2] we have studied the composites consisting of high T_c - superconducting material with magnetic fluids (fine magnetic particles dispersed in liquid carrier). The structuralization effects of high T_c - superconductive particles were observed using the Skjeltorp's idea [3] of magnetic holes. In paper [4] using the effective medium theory the influence of magnetic surrounding and structure of high T_c - superconductive particles on the critical magnetic fields of a superconductor has been studied. The magnetic flux exclusion properties of superconductive material are strongly affected by the presence of magnetic field larger than the lower initial field H_{c1}. As an applied field rises above H_{c1}, magnetic flux penetrates into the body of the sample, reducing the amount of excluded flux. Many of the new high T_c - superconducting materials have low H_{c1} values and thus are especially sensitive to magnetic fields. Flippen [5] studied the magnetic flux exclusion properties of composites of ferromagnetic and high T_c - superconductive powder. It was shown that the magnetic state of a magnetic material affects the flux exclussion properties of a superconductive material.

Our aim was to study the flux exclusion properties of composite material made up of high T_c - superconductive $YBa_2Cu_3O_{7-x}$ - oxide and magnetic particles (Fe_3O_4) traditionally used in magnetic fluid technology.

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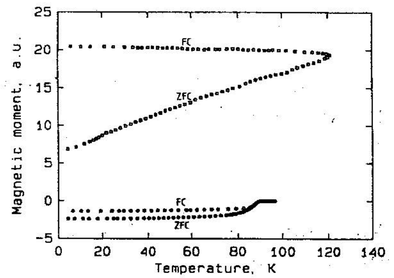

Fig. 1. ZFC and FC curves for magnetite powder (squares) and for $YBa_2Cu_3O_{7-x}$ powder (circles).

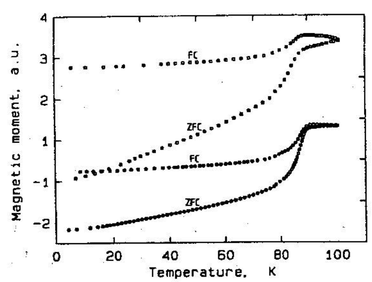

Fig. 2. ZFC and FC curves for two mass concentrations of magnetite: 5.18% - circles and 20.19% - squares respectively.

2. Experimental details

The samples of high T_c - superconductive powder of $YBa_2Cu_3O_{7-x}$ - oxide were prepared by standard sol - gel technique using the autocatalytic pyrolitic decomposition of yttrium, barium and copper nitrates sol [6]. The interproduct was annealed at 920°C in air for 16 hours and sintered at 920°C in oxygen for 16 hours in a platinum boat and cooled at a rate of 20°C/min. Morphology observation of this product by using scanning electron microscopy showed that grains of 0.4 μm in size are stuck together in aggregates of 5 – 15 μm in size. The magnetite particles were prepared by a standard technique from ferrous and ferric salts. The electron microscopy examination showed magnetite particles with the mean diameter D_v = 10 nm and a standard deviation σ = 0.33. Composites of the above mentioned powders were made by thoroughly mixing appropriate masses of the two in a nonreactive fluid such as ethanol, followed by evaporating fluid and pressing finally. Measurements of magnetic and magnetic flux exclusion vs. temperature were carried out using standard VSM-magnetometer. The results were obtained by

1. zero field cooling (ZFC) the sample to 4.2 K, after which the desired field was applied. This ZFC-curve corresponds to the shielding effect,
2. field cooling (FC) the sample in the applied field, thereby measuring the flux exclussion corresponding to a true Meissner effect. The magnetic field $\mu_o H$ = 2 mT was used in measurements.

3. Results and discussion

Figure 1 shows ZFC and FC curves for pure magnetite and $YBa_2Cu_3O_{7-x}$ oxide used in the experiments. Figure 2 represents ZFC and FC curves for two mass concentrations of magnetite. The very nice "pigeon-like" behaviour is observed for concentration c = 20.19%. In Fig. 3a is plotted difference of Meissner (shielding) effect and linear combination of pure magnetite and superconducting signal as a function of mass concentration of magnetite at temperature T = 25 K. The curves have minimum at a

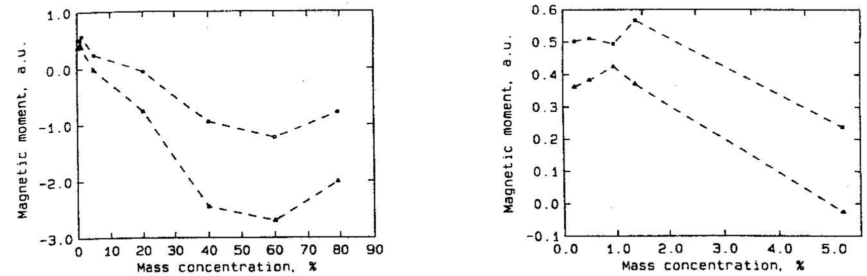

Fig. 3. (a) The difference between Meissner effect of the whole composite and linear combination of pure magnetite and superconductor signal (triangles); the same for shielding effect (squares) as a function of mass concentration of magnetite particles. (b) The same as in Fig. 3a but in low concentration range.

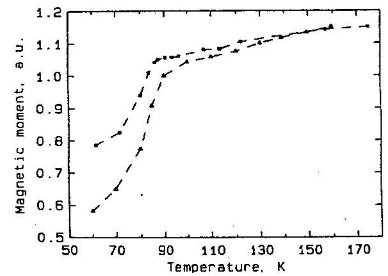

Fig. 4. The influence of magnetic state of magnetite on Meissner effect (triangles - demagnetized sample, squares - sample magnetized in field $\mu_o H = 0.4T$). Mass concentration of magnetite particles is 0.211%

concentration of about $c \simeq 60\%$. Very interesting behaviour is observed in low concentration range. This is shown on Fig. 3b. Fig. 4 shows an example how the magnetized magnetite (magnetized at $\mu_o H = 0.4\ T$) effectively shifts the flux exclusion properties. Our initial attempt to explain the measured dependences of composite materials properties was based on the effective medium theory principles. The effective permeability μ_{ef} of two component composite system has been calculated using known permeabilites of superconductive μ_s and magnetic μ_f parts by means of the often used Bruggeman's formula [7]

$$\frac{\mu_{ef} - \mu_S}{\mu_f - \mu_s} \left(\frac{\mu_f}{\mu_{ef}}\right)^{1/3} = 1 - \varphi, \qquad (1)$$

where φ is a volume fraction of magnetic particles in composite. But even preliminary comparison of measurements leads to significant differences. Therefore, the purpose of further work was to abandon effective medium theory in such systems and to investigate the observed anomalies at low concentration region of magnetic particles. Here the presence of magnatic particles seems to have a significant influence on Meissner effect. Let us assume that system of magnetic particles in composite can be considered as a system of randomly positioned magnetic dipoles with density n, with Gaussian distribution of magnetic fields [8]

$$P(\vec{H}) = \frac{1}{2\delta\sqrt{\pi}} \exp\left[-\frac{H^2}{4\delta^2}\right] \tag{2}$$

with Gaussian spread

$$\delta = \sqrt{\frac{4\pi p^2 n}{9a^3}}, \; p = \frac{4}{3}\pi a^3 M_s \,, \; n = \frac{\varphi}{\left(\frac{4}{3}\pi a^3\right)} \,, \tag{3}$$

dependent on the magnetic particles diameter a, their magnetic moment p and saturation magnetization M_s. Then the probabillity P_{c1} the local field intensity $|\vec{H}|$ at a certain randomly choosen position in composite is larger than the lower transition field of superconductor fraction H_{c1} is

$$P_{c1} = 4\pi \int_{H_{c1}}^{\infty} P(\vec{H})\, H^2\, dH = 1 - \frac{2}{\sqrt{\pi}} \Gamma\left[3/2, 0, \frac{H_{c1}^2}{4\delta^2}\right] . \tag{4}$$

In the framework of mean field approximation we can assume that the introduced P_{c1} is close to a volume fraction of normal and mixed-state regions in the second order superconductor. To demonstrate this assumption for typical composite parameters quantitatively we choose the probability level 0.99, and therefore start analysing inequality

$$P_{c1} > 0.99 \,, \tag{5}$$

which is equivalent to

$$H_{c1} < 0.47923\,\delta \simeq 1.159\, M_s \sqrt{\varphi} \tag{6}$$

Taking $\mu_0\, M_s = 0.4\; T$ and $\mu_0\, H_{c1} = 0.04\; T$ and using (6) we can see, that even a very small magnetic particles admixture with $\varphi > 0.0074$ is high enough for a significant Meissner effect disturbance (5). This probably corresponds qualitatively to observed magnetic anomalies in the low concentration region and recovers a source of difficulties in our inital application of effective medium theory.

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