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AC-susceptibility of $YBa_2Cu_3O_{7-y}$ ceramics in terms of the percolation theory

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An approach is formulated which permits to analyze AC-susceptibility data at various amplitudes h in terms of the percolation theory. The relative superconducting volume $v(T)$ and the fraction of superconducting contacts $p(T, h)$ are the main variables of the problem. The whole set of curves $\chi(T)$ at various h is situated between two limiting curves which correspond to small enough and large enough amplitudes h. The function $v(T)$ can be obtained from the second limiting curve. Its derivative is the T_c-distribution function with reliably observable width and structure. By changing the oxygen content this distribution function can be smoothly shifted along the T-axis. The function $p(T, h)$ at $h\neq 0$ depends on a definite combination of T and h: $p=p(\gamma)$ where $\gamma=h/H_0(1-T/T_0)$. The parameter $p(T, h=0)$ turned to depend on the oxygen content as well. It seems to increase up to $p\approx 1$ when T_c is near 60 K. This is one of the experimental motivations to extend the meaning of the parameter p so that it would take into account cooperative phenomena such as long-range coherent state along superconducting clusters.

1. Introduction

Contactless measurements of the surface impedance, which in the case of superconductors are widely known as AC-susceptibility measurements, serve as a very convenient and widespread method of study of high-T_c superconductors (see, e.g., refs. [1–6]). Therefore, it is very important to realize how these measurements depend on the material inhomogeneity, percolating structure of the superconducting clusters, presence of pores and extended normal inclusions, intergrain weak links, etc. In this paper such a study has been accomplished for the $YBa_2Cu_3O_{7-y}$ ceramics.

In describing the processes which determine the ceramics' impedance, the critical state model in some form (e.g., ref. [7]) or the weakly connected loop model [8] are usually used. In the present paper an attempt is made to examine the experimental results within the framework of the percolation theory [9,10] using the peculiar properties of the superconducting state and Josephson junctions for setting up parameters of the percolation problem. Such an approach originated from the observation [11] according to which the real part of AC-susceptibility $\chi(T)$ for broad transitions had a kink always at one and the same χ value. This constancy of the kink level can be rather naturally interpreted in terms of screening properties of finite superconducting clusters.

We suppose a grain to be an element of the superconducting network. Its average size in $YBa_2Cu_3O_{7-y}$ ceramics, $d\approx 1$ µm, is much larger than the magnetic penetration depth, $\lambda=1500$ Å, and the coherence length $\xi=40$ Å. Due to inequality

$$d\gg \xi, \lambda \tag{1}$$

we can assume that the superconducting transition of a definite grain does not depend on the state of surrounding grains. The grains themselves are supposed to be ideal, i.e, they have a sharp transition at a certain temperature, T_c, which, however, may be different for various grains. To describe the superconducting transition in a granular medium the concept of two-stepped transition is often used: an intragrain transition occurs at temperature T_c but superconducting wave functions of different grains remain out of phase down to $T_0<T_c$, when a long-range phase ordering takes place [12–14]. Anyway, the knowledge of percolation parameters is neces-

sary to provide a consistent analysis of such kind of cooperative phenomena [15,16]. Therefore, we consider that it is possible and expedient to start with an attempt to analyze the experimental data taking into account cooperative phenomena only formally, if at all.

The paper is organized as follows:

The details of experiments and experimental data as the primary material for the following analysis, are presented in section 2. All the measurements were carried out with one ceramic sample, the oxygen being gradually removed from it. This means that all the experiments were performed on the background of the same geometrical structure – some fixed network of grains. Each annealing leads to a new distribution of the oxygen content over the grain assembly, that is to a new distribution function $g(T_c)$ of the superconducting transition temperatures.

The main content of the paper is concentrated in section 3 where an attempt is made to interpret consecutively the experimental data on the basis of percolation conceptions. As a subject to the percolation theory, the $YBa_2Cu_3O_{7-y}$ ceramics presents a combined problem: superconducting (s-) or normal (n-) may be both grains – "sites", and junctions between them – "bonds". Accordingly, one can introduce two parameters: v – the fraction of s-site or, in other terms, the s-part of the sample volume

$$v(T)=\int_T g(\tau)\,d\tau\,, \qquad (2)$$

and p which is the fraction of open bonds between s-sites (an s-bond is an open bond and an n-bond is closed). The merit of AC contactless measurements is that by increasing the amplitude of the alternating field, h, they permit closing the bonds, without changing the value of v. In particular, with high enough amplitudes, it is possible to decouple all grains, i.e, to destroy all s-bonds, and to obtain $p=0$. Under these conditions the total volume of s-grains, i.e., function $v(T)$ can be measured.

Using function $v(T)$ as a fundamental quantity, we propose in section 3.2 a model of overlapping diamagnetic rings and use it to describe the AC-susceptibility at low h. In section 3.3 we apply the same model to interpret the data with intermediate h values.

In the final part, section 4, we discuss once again whether inequality (1) is sufficient to apply the percolation theory and what features of the AC-susceptibility of the $YBa_2Cu_3O_{7-y}$ ceramics can be ascribed to manifestations of cooperative phenomena.

2. Experimental

A cylindrical sample of 2 mm in diameter and about 10 mm in height was placed inside one of the two identical couples of induction coils connected towards each other. The coils were disposed in liquid helium over the external surface of a small overturned dewar while the sample together with a thermometer and a heater were placed inside the dewar. To prevent temperature gradients, the sample was placed in a sapphire container.

The sample was made of ceramics, its density being 5.6 g/cm^3. X-ray examination did not detect any traces of extraneous phases in this material. The sample in the initial state had a sharp superconducting transition – see curve "0" in fig. 1. After a series of measurements at different frequencies and amplitudes it was subjected to vacuum annealing, which resulted in a slight decrease in the oxygen content. Then a new series of the low-temperature measurements, followed by further annealing, was done and so on.

The experiment consisted in measuring the disbalance signal by a lock-in amplifier, i.e., in the measurement of the sample linear response at various frequencies ω and amplitudes h of the stimulant field. The coefficient of nonlinear distortions has not been measured specially.

The measurements were carried out in the frequency range of 10^2–10^5 Hz. The amplitude of alternating field h varied from 2×10^{-2} to 40 Oe. The use of various frequencies was caused by methodical reasons only. Higher frequencies were used at low amplitudes to increase sensitivity. heating of the sample and thermometer by alternating currents induced to use lower frequencies at high amplitudes. Though the frequency dependence of the signal does exist in this range, it is considerably weaker than the effects that will be discussed below. However, for each amplitude h, there exists some frequency ω_h below which the $\chi(\omega)$ dependence practically disappears. All our results refer to the frequency range $\omega<\omega_h$. In particular, at extremely low $h \leq 10^{-2}$ Oe there was no frequency dependence up to 10^5 Hz.

Phase-sensitive detection permitted us to measure both components of the signal simultaneously. In this paper we discuss only the real part of AC-susceptibility, i.e, the imaginary part of surface impedance. The value of Y, depicted on the ordinate axis in figs. 1–3, is proportional to magnetic moment of the sample

$$Y = -\frac{4\pi\chi(1-n)}{1+4\pi\chi n}. \tag{3}$$

Here n is the demagnetization factor of the sample (in our case $n \approx 0.05$) and χ means an effective value of susceptibility, resulting from the averaging over the volume of the sample. If $n \ll 1$, then Y is in the first approximation normalized susceptibility

$$Y \approx (-4\pi)\chi. \tag{4}$$

The value of $Y=1$ means that the magnetic flux is fully extruded from the volume of the sample. Fig. 1 presents $Y(T)$ curves, obtained at extremely low amplitudes h, when neither tenfold change of the amplitude h, nor the tenfold frequency change affects the signal. For convenience the curves are numbered. A digit near curves $i=0, 1, ..., 5$ means the number of vacuum annealings the sample has been subjected to. On all the curves, except for the initial one $i=0$, there exists a kink at low values of Y. One can see this kink also in all the papers, where the corresponding experimental examples are presented. It arises at a temperature, when the concentration of the s-grains becomes so large that inside the clusters, there appear extended n-regions shielded from an external field [11]. Similar explanations have been already proposed previously [2,4]. For our approach dealing with percolation ideas, it is important that the kink is always located at the same height, at $Y \approx 0.1$.

It is well known at AC-susceptibility of ceramics depends on h [1–6]. Fig. 2 depicts, for example, two whole sets of $Y_i(T)$ curves, obtained after the third and the fourth annealings of the sample ($i=3, 4$). The main result here is an obvious existence of two limiting curves: $Y^{(1)}(T)$ at low h (upper curve) and $Y^{(2)}(T)$ at high h (lower curve). Similar sets of

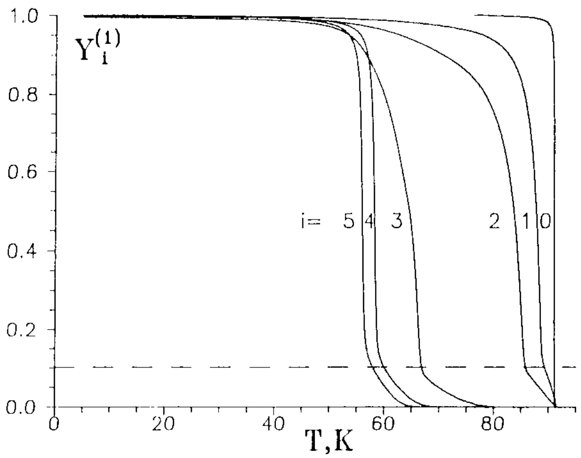

Fig. 1. Curves $Y_i^{(1)}(T)$ at extremely small amplitudes h. $\omega/2\pi = 10^5$ Hz. Digits near curves mean the number of vacuum annealings. Dashed line marks the level of the kink.

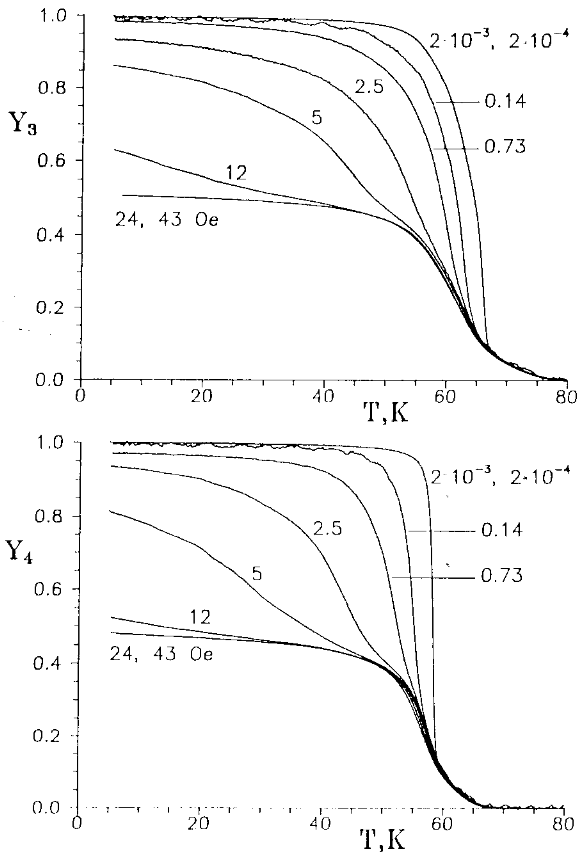

Fig. 2. Sets of curves $Y_3(T)$ and $Y_4(T)$ at different h. The curves are labelled by h values. The frequency is 100 Hz anywhere except the upper limiting curves.

curves were obtained for all other i. All upper limiting curves are shown in fig. 1. All $Y_i^{(2)}(T)$ curves will be given below in fig. 3. Note that gradual formation of lower limiting curve with increasing h can be found in the experimental data presented in papers [1,3,4,6].

3. Interpretation of experimental data

Consider the following assumptions:

(1) Oxygen deficiency ($0<y<0.5$) leads to a variety of T_c values for different grains, though the transition of each grain remains narrow. The arguments in favor of such assumption are based on the following [17]. The oxygen migration through the grain boundaries is too complicated and oxygen concentration leveling takes place inside grains only. The grains are supposed to be too small for domains of ortho-I and ortho-II phases to form inside them. Small size persuades the grains to have a homogeneous oxygen distribution with intermediate concentration values and, as a result, with intermediate transition temperatures, $T_c<90$ K.

(2) Even at our highest amplitudes, h, the alternating field does not destroy superconductivity of a grain, i.e., v depends on T only, but not on h: $v=v(T)$.

(3) Field h can destroy s-bonds. On the upper limiting curve $Y^{(1)}(T)$ this process does not take place because of the smallness of h, so that one has $p_{\max}(T)$ due to the inner nature of the sample. And vice versa, on the lower limiting curve $Y^{(2)}(T)$, all s-bonds are destroyed and $p=0$. Note that to the intermediate curves $Y(T)$ some p values may be attributed only formally. The field destroys s-bonds not randomly but depending on realized structure of s-sites, at those regions where the density of shielding currents is the largest. Therefore, p stops to be an independent parameter, though an average number of s-bonds formally exists: $0<p<p_{\max}$.

3.1. The lower limiting curve

In a strong AC field, i.e., with all the bonds closed, a sample is a mixture of n-grains with $\chi=0$ and s-grains with $\chi=-1/4\pi$. The concentration v of the s-phase should be deduced from the measured $Y^{(2)}$ value. In the mean-field approximation

$$\chi=(-1/4\pi)\frac{3v_{\text{eff}}}{2+v_{\text{eff}}}. \tag{5}$$

Here v_{eff} is an effective concentration of s-phase, which differs a little from v because of the finite value of penetration depth of magnetic field into grains. Substituting eq. (5) into (3) we obtain

$$v_{\text{eff}}=\frac{2Y^*}{3-Y^*}, \qquad Y^*=Y(1-n+Yn)^{-1}. \tag{6}$$

The right expression takes into account the demagnetization factor of the sample; the left one, in the mean-field approximation, the demagnetization factor of grains. Besides expression (2) for v, we have for v_{eff}

$$v_{\text{eff}}(T)=\int_t m\left(\frac{\lambda(T/\tau)}{d}\right)g(\tau)\,\mathrm{d}\tau, \tag{7}$$

where $m(\lambda/d)$ is the normalized magnetic moment of an s-grain depending on the λ/d ratio. It depends on the grain shape, but in the asymptotic expressions

$$m(\lambda/d)=\begin{cases}1-\alpha_1(\lambda/d), & \lambda/d\ll 1,\\ \alpha_2(\lambda/d)^{-2}, & \lambda/d\gg 1,\end{cases} \tag{8}$$

this dependence reduces to the values of coefficients α_1 and α_2 only [19]. For a sphere of radius d coefficients $\alpha_1=3$ and $\alpha_2=\frac{1}{15}$.

According to the superconducting theory, in the vicinity of T_c

$$(\lambda_0/\lambda(T))^2=\beta(T_c-T)/T_c, \qquad \lambda_0\equiv\lambda(0). \tag{9}$$

The experimental value of the coefficient $\beta\approx 3$ [20,21]. As λ^{-2} approaches zero at T_c linearly, when $\lambda_0/d\ll 1$ and the distribution function $g(T)$ is broad enough,

$$(\mathrm{d}v/\mathrm{d}T)^{-1}\gg T(\lambda_0/d)^2, \tag{10}$$

the difference between v and v_{eff} reduces to two corrections, small in parameter λ_0/d,

$$v(T)\approx v_{\text{eff}}\left(1+\alpha_1\frac{\lambda_0}{d}\right)-\frac{\mathrm{d}v_{\text{eff}}}{\mathrm{d}T}\frac{T}{2\beta\alpha_2}\left(\frac{\lambda_0}{d}\right)^2. \tag{11}$$

Condition (10) means that the width of the distribution function is larger than the temperature interval, where the penetration depth of the field into a grain, $\lambda_0(1-T/T_c)^{-1/2}$, is comparable with d. This condition is obviously violated for the sample in the initial state, not subjected to vacuum annealing yet ($i=0$). We used in this case another limiting procedure, substituting the δ function into eq. (7):

$$g(T)=0.9\delta(T-T_c) .$$

Here the factor 0.9 is the ratio of the sample density to the ideal one. Having used the $\lambda(T)$ dependence (9) in the vicinity of T_c, with the experimental value for β, we obtained $d/\lambda_0\approx 12$ from the curve $Y_0^{(2)}(T)$. The value obtained is in a good agreement with preliminary estimates. Thus justifies the use of the δ function.

Now while handling all the other curves with the help of expressions (6), (11), we may use the not estimated but experimentally measured value of λ_0/d. Figure 3 presents the results of such processing together with the initial $Y_i^{(2)}(T)$ curves.

It seems rather strange, the limiting values of functions $v_i(0)$ decrease from one annealing to another but at the same time the distribution functions $g_i(T)$ have no low-temperature tails. It looks as if one part of grains decreases their T_c with annealing in a correlated manner, while the others become abruptly unobservable (normal, or break down into many small grains transparent to the field, or become permeable to vortexes, etc.), and as if the volume of this second group increases with every annealing. At the same time, X-ray examination did not detect any traces of the tetra phase in states $i=4$, 5. We have no explanation of this fact yet. We will discuss this question once more in the final part of the paper.

Below, we consider corrections $(Y^{(2)}-v)$ to be small in accordance with fig. 3, i.e., $Y^{(2)}\approx v$.

3.2. The upper limiting curve

Let the normal conductivity of the medium be so small that exponential decay of the field can be neglected. Let us introduce the distribution function of loops formed by grains with respect to the areas of these loops. Suppose the number of loops with area s in the interval ds per unit volume is $\varphi_{v,p}(s)\,ds$, and the volume, shielded by these loops is

$$f_{v,p}(s)\,ds=s^{3/2}\varphi_{v,p}(s)\,ds . \tag{12}$$

If $F_{v,p}(s)$ is a fraction of volume, shielded by all s-grains and by all loops with the areas less than s, overlapping taken into account, then

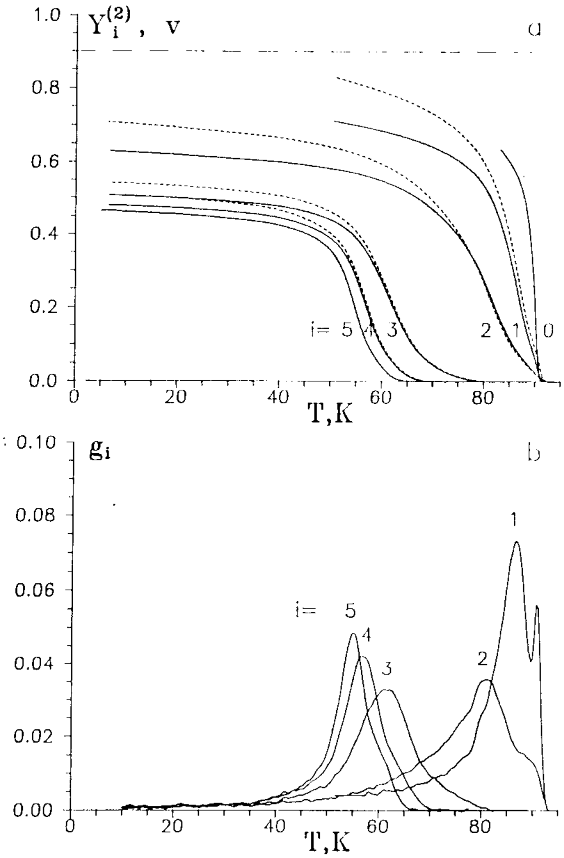

Fig. 3. (a) Limiting curves $Y_i^{(2)}(T)$ (solid lines) and the volume of s-phase $v_i(T)$ (dashed lines). $v_i(T)$ are obtained under assumption $d/\lambda_0=\text{const}=12$. (b) Distribution function of s-grains over the transition temperatures $g_i(T)=dv_i/dT$.

$$[1-F_{v,p}(s)]f_{v,p}(s)\,ds=dF_{v,p}(s) , \tag{13}$$

where from

$$F_{v,p}(s)=1-(1-v)\exp(-I(s)) ,$$
$$I(s)=\int_0^s f_{v,p}(s)\,ds , \tag{14}$$

and at the upper limiting curve $Y^{(1)}(T)$ one has

$$Y^{(1)}(T)=F_{v,p}(\infty)$$
$$=1-(1-v)\exp(-I(\infty)) . \tag{15}$$

It is seen from eqs. (14) and (15) that $\exp(-I)$ is the mean value of the magnetic permeability $\bar{\mu}=\overline{(1+4\pi\chi)}$ over the space between s-grains.

All the functions introduced, φ, f and F depend on p and v values. It is easy enough to deduce them explicitly when $v=p=1$. Indeed, suppose a 3D lattice in which each site has z neighbors, and a particle making a random walk on such a lattice. The number of paths by which it can get back after N steps is $z^N N^{-1.5}$. If one takes into account the self-avoided paths only, then their number is $(\tilde{z})^N N^{-1.8}$ [22]. For a simple cubic lattice $z=6$, and $\tilde{z}=4.7$ [22].

After normalizing the number of paths per lattice site, the number of loops containing N grains is

$$\varphi_{1,1}(N) \propto (\tilde{z})^N N^{-2.8} . \tag{16}$$

Hence, putting $s \propto N^2$, one finds

$$\varphi_{1,1}(s) \propto (\tilde{z})^{\sqrt{s}} s^{-1.4} . \tag{17}$$

Probably expression (17) may be generalized to the case of an almost regular lattice, $p \approx 1$, $v \approx 1$ if one substitutes $\tilde{z}$ by $pv\tilde{z}$. However it is certainly unapplicable for small p or v in the vicinity of the percolation threshold and below it, because it does not take into account the cluster pattern of grain distribution. Instead, the problem of the ant in the labyrinth must be dealt with [9].

An attempt trying to obtain information about φ and f functions right from the experiment is quite natural. For this purpose fig. 4 depicts $I^2(v)$ functions, where

$$I = \ln[(1-Y_i^{(1)})/(1-Y_i^{(2)})] ,$$

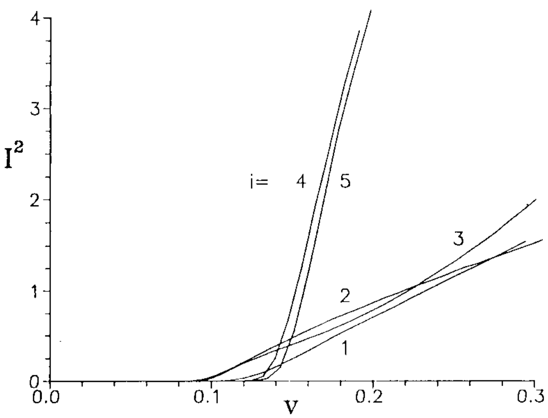

Fig. 4. Dependences of integral (14) on the volume of the s-phase for realizations 1–5.

for $i=1, ..., 5$. Two things can be deduced from the figure.

(a) It seems that the dependence $I(v)$ can be described by the power law

$$I = K(p)(v-v_0)^{1/2} . \tag{18}$$

Coefficient $K \approx 10$ for realizations $i=4$ and $i=5$ with narrow distribution function, $T_c \approx 60$ K and, apparently, with $p \approx 1$. Other realizations have $K \approx 3$.

(b) The fact that I is not a universal function of v means that the parameter p differs for various realizations. A sharper increase of I with v points to a greater value of p. Intersection of curves $Y_3^{(1)}$ and $Y_4^{(1)}$ in fig. 1 leads to the same conclusion about the ratio of p values. Compare with fig. 3, where $p=0$ and where $Y_3^{(2)} > Y_4^{(2)}$ in the whole temperature range.

3.3. Intermediate curves

The amplitude of the AC field influences the response of the $YBa_2Cu_3O_{7-y}$ ceramics because s-junctions become transparent to the field. It is easy to conclude from fig. 2 that s-bonds are not destroyed simultaneously. There are two reasons for this.

First, the junctions are situated in different geometrical conditions. Some of them are shielded by the neighboring s-clusters, while the others are in the regions of field concentration. The field amplitude on the junction may thus be both, less and more than h. Second, there is a variety of parameters of the junctions itself, described by the variety of critical currents $I_c(T)$ and coupling energies

$$E = I_c\phi_0/2\pi c, \qquad \phi_0 = \pi\hbar c/e . \tag{19}$$

The two causes of dispersion of the junctions response to the field should be taken into account in integral (14) in different ways. Geometrical dispersion can be accounted for by making the upper limit s of the integral to depend on h, while the variety of junctions should affect the integrand. However, we do not see a way yet to distinguish these two sources of inhomogeneity from the experiment. Therefore, in this section we describe the observed dependences from the phenomenological point of view.

As is seen from fig. 3, the intermediate curves separate from the lower limiting curve one by one, at quite definite temperatures. These are the temperatures, when the strongest (or the most shielded) bonds cannot withstand the applied field h any further. Denoting the corresponding field value h by h_c we summarized all the data on $h_c(T)$ dependences for various realizations $i=1, ..., 5$ in fig. 5. In all the cases straight lines were obtained. Introducing two characteristic constants h_{0i} and T_{0i}, we obtain

$$h_c = h_{0i}(1 - T/T_{0i}) . \tag{20}$$

(Maybe it would be more correct if a value of the mean field $h(1-Y)$ or of the local field $h(1-Y/3)$ will stand for h_c and not that of h, the result being still independent of this.)

The linear dependence (20) may be the result of the T-dependence of the critical current in the vicinity of T_c

$$I_c \propto \Delta\,\mathrm{th}(\Delta/2T) \propto \Delta^2 \propto (T_c - T) \tag{21}$$

(Δ is the superconducting gap).

It is natural that in this case T_{0i} would be close to the temperature at which the maximum of distribution function $g_i(T)$ is observed. It is easy to check that it is really so (compare figs. 3 and 5).

Now we will try to find a universal form for all intermediate curves. Let us start with model (12)–(15), where the susceptibility depends on the loops distribution function. Consider rather low temperatures, where $v(T) \approx$ const. In this region the function $f_{v,p}$ and, as a result, χ both change mainly because of the dependence of p on h. Then it seems natural that the field h normalized by the critical value h_c from eq. (20) is taken as an argument of the universal function we are looking for: experimentally measured temperature dependence can be transformed into a function of h/h_c due to the T-dependence of h_c. So as an argument we use the quantity

$$x = \frac{h}{h_{0i}(1 - T/T_{0i})} = \frac{T_{0i} - T^*_{hi}}{T_{0i} - T} , \tag{22}$$

where T^*_{hi} is the temperature at which curve $Y_i(T)$, obtained with the amplitude h, separates from the lower limiting curve $Y_i^{(2)}(T)$. Point $x=1$ corresponds to this separation temperature: $T = T^*_{hi}$.

A function of this new argument can be given by the

$$\bar{\mu} = (1 - Y_i)/(1 - Y_i^{(2)}) . \tag{23}$$

According to eq. (14) it is equal to $\exp(-I)$. Besides the dependence on h/h_c, it contains in the integrand only parameter v which is constant in the temperature range under consideration.

In fig. 6 all the intermediate curves with different i are plotted in these coordinates (some of the curves are not shown not to overload the graph). It is not surprising that all the curves converge at the point of $p = x = 1$. This results from the definition: at the separation point $Y_i = Y_i^{(2)}$. More informative is the left part of the graph, where all the curves transform into straight lines

$$\bar{\mu} = \gamma_i x , \tag{24}$$

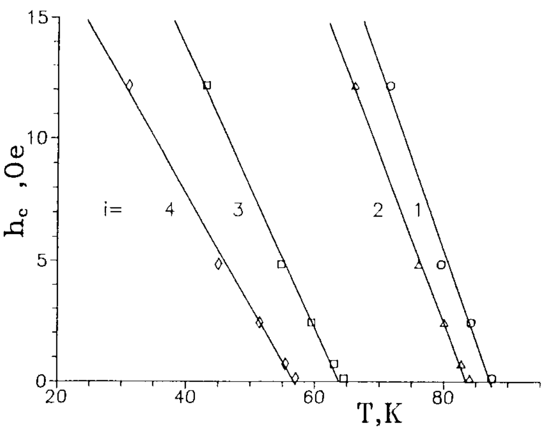

Fig. 5. Separating field h_c against the temperature for different realizations.

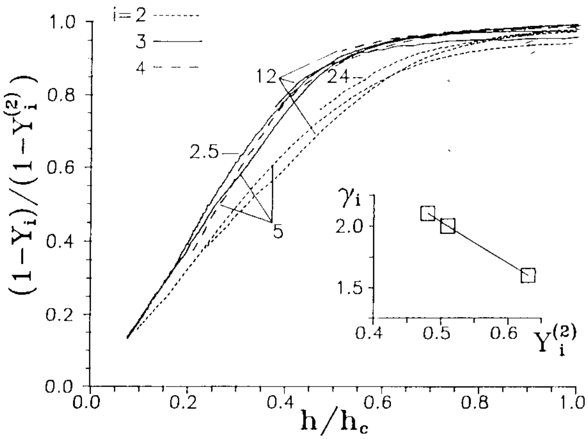

Fig. 6. Universal dependence of the mean magnetic permeability in the space off the grains on the normalized amplitude h. Inset shows the change of the slope of the lines (24).

whose slopes depend on i, but not on h. The fact that the lines (24) pass through the origin means, according to eq. (14), that at low h the full shielding of the volume by s-currents takes place. The same follows from the limiting curves $Y^{(1)}$ in fig. 1.

It should be noted that in essence transformation (22), (23) is similar to another one which was proposed in ref. [6] on the basis of other considerations. Therefore, it is not an argument in favor of the model of overlapping diamagnetic loops (12)–(15). This model could be checked by theoretical analysis of the dependence of γ_i on $v(0)$. Apart from this, it should be yet elucidated why a linear dependence (24) takes place in such a wide range of h values. However, both steps require mathematical elaboration of this model.

4. Discussion: results and problems

Two alternative approaches to the experimental material of this paper are possible – from the side of the percolation theory and from that of physics of superconductivity. Consider the first one:

A problem exists, to what extent the percolation theory in its classical form is suitable for description of superconducting transition in the inhomogeneous medium and whether inequality (1) is a sufficient condition for its usage. This question in turn falls into two.

First, model (12)–(15) implicitly suggests that the mean field h in our s-composite does not decay inside the sample. This assumption fits the experimental fact that at the percolation threshold the screening factor is $Y^{(1)} \approx 0.6$–0.7 [1,17], though the size of the sample is usually by three orders of magnitude larger than the average size d of the structure elements (grains). Even if an exponential decay of the field inward the sample exists, then in the vicinity of the threshold an effective penetration depth is $\delta_{eff} \gg d$. Not in any case, neither in ours nor in earlier experiments [1–6] any peculiarities of function χ at the threshold were observed.

The problem is not in the field distribution controlled by the critical currents [7,23] but in the propagation of an incident electromagnetic wave of an infinitely small amplitude through the s-composite space. (We mean s-clusters in an insulating medium with elements which fit condition (1) and without dissipation in junctions. Apparently, the experiments with $YBa_2Cu_3O_{7-y}$ at small amplitudes h simulate just such medium.) Sometimes, in discussing the AC-susceptibility of s-composites, an exponential decay of the field is introduced as something selfevident. However, the AC-response of composite superconductors has been examined theoretically, as far as we know, only under the condition $d < \lambda$ (see [24] and references therein).

Second, we have not discussed yet the possible contribution of cooperative phenomena. In a composite apart from the temperature of the superconducting grain transition, T_c, there should be another characteristic temperature T_0 of the long-range phase ordering [12–14]. In general, this may be included into our model by introducing sharp change of p at point T_0. Maybe, the kink in the $Y^{(1)}(T)$ curves is due to just this process, and not to the cluster shielding, described above in classical terms. This could be stated more definitely, if we had calculations of shielded volume $\chi(v)$ even under the simplest assumptions – without correlation effects and with $p=1$.

Let us try now another approach, along the lines of the physics of superconductivity. The presented data prove that it is possible to move T_c gradually from 90 to 60 K, by decreasing the oxygen content. The absence among $g_i(T)$ functions in fig. 3 of one with a maximum between 65 and 80 K is a casual factor, caused by the conditions of the third annealing. In another set of annealings, such functions existed [11].

In general, existence of parameter λ/d gives some freedom in the transition from $Y^{(2)}$ to v. Namely, one can choose for each i its own λ/d value, keeping the value $v(0)$ constant: $v(0)=0.9$. For the $i=4$ curve this would demand the value of d/λ_0 to be about 5 instead of 12. The result of such processing is depicted in fig. 7. A comparison of figs. 3 and 7 shows that the positions of maxima of $g_i(t)$ functions have not changed.

In fig. 7 the area under all $g_i(T)$ curves is the same: $v(0)=0.9$ and the difficulty in the interpretation which we have discussed above in section 3.1, vanishes. However, another, a more serious one appears instead.

The increase of the λ_0/d parameter lifts the func-

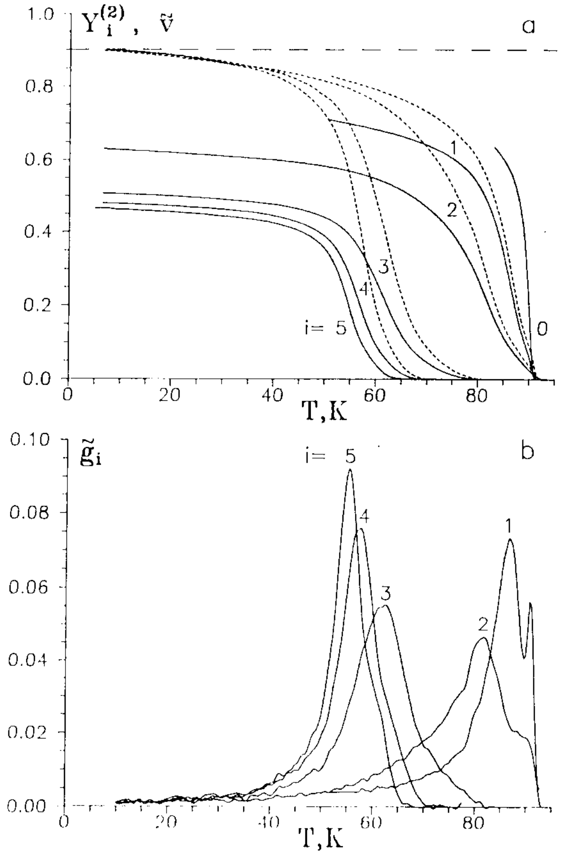

Fig. 7. The same as in fig. 3 but curves $\tilde{v}_i(T)$ are obtained under an assumption $\tilde{v}_i(0) = \text{const} = 0.9$ ($\tilde{g}_i = d\tilde{v}_i/dT$).

tion $v(T)$ in the whole temperature range and, in particular, in the vicinity of the kink

$$Y^{(1)} = Y^{(2)} \approx 0.1 \ .$$

This leads to inequality $v > Y$ and the difference increases with i. We would have to give up the statement that the kink in $Y^{(1)}(T)$ curves is always observed at one and the same fraction of the s-phase, a statement which is natural in the framework of percolation theory. And we would have to explain appearance of the kink at one and the same Y value instead. This again brings us to the problem of the coherent state.

In section 3.2 we have already mentioned that the function $I(v)$ at low h is not universal and that p is larger for the realizations with $T_c \approx 60$ K. Of course, the explanation may be just a structural one – the boundaries between the grains with intermediate oxygen concentrations $0.5 < y < 1$ may be wider and less regular than at $y \approx 0.5$. However, another explanation is possible, that right here the cooperative phenomena become important. It follows from fig. 3, as well as from fig. 7, that realizations $i = 4$, 5 with $T_c \approx 60$ K have much narrower distribution functions. The problem is whether this may be important, i.e., whether the variety of the grain's T_c may affect the appearance of a coherent state. Instead of convincing theoretical considerations, we can propose only an analogy. The key idea of Anderson's model of the metal–insulator transition in disordered media is that about the chains of resonant sites. The probability for two neighboring sites to be resonant depends on the dimensionless ratio of the band width W to the overlap integral J. The first controls the mean energy difference between these two sites, the second determines the difference permitting the sites to remain resonant. In our case the width of the distribution function g^{-1} is a natural counterpart to the bandwidth W, and the Josephson energy E defined in eq. (19) can be proposed to the role of J. It follows from fig. 5 that E almost does not change from one realization to another.

There exists one experimental argument pointing to the cooperative phenomena as to a possible cause of the sharp increase of functions $Y_4^{(1)}$ and $Y_5^{(1)}$. The structural causes for the contrast between them and functions $Y_2^{(1)}$ and $Y_3^{(1)}$ (the width of boundaries, etc.) would remain in the magnetic field. But, according to section 3.3, the differences vanish with increasing h: curves $\bar{\mu}(x)$ for realizations $i = 3$ and $i = 4$ coincide. This argument, however, may hardly be considered as a decisive one.

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