

# Penetration of low magnetic fields into ceramic HTSC (low-field electrodynamics)

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We investigate theoretically and experimentally the penetration of magnetic fields much weaker than the granule critical field into HTSC ceramics. Expressions are obtained, using the theory of the critical state of a random Josephson medium in low-field electrodynamics, for the harmonics of the induction  $\bar{B}(t)$  as functions of the external parameters, in two asymptotic cases—low field and thin sample. For a thin sample, in particular, it is shown that the harmonics are oscillatory functions of the alternating-field amplitude. The behavior of the linear and nonlinear susceptibilities (in an alternating field) and of the hysteresis loop and of the magnetization curve (in a constant field) are investigated experimentally in a wide range of temperatures and fields. The experimental and theoretical results are in satisfactory agreement. A number of quantities descriptive of the low-field electrodynamics of HTSC, and their behavior as functions of temperature, are determined. An explicit form of the function  $f_c(H, T) = f_0(T)H_0^2(T)/(H^2 + H_0^2(T))$  is determined experimentally.

## 1. INTRODUCTION

Many recent publications deal with the penetration of magnetic fields much weaker than the first critical field of the granules themselves into ceramic high-temperature superconductors (HTSC). This phenomenon, called low-field electrodynamics, arises because ceramic HTSC is a random Josephson medium that behaves as a standard type-II superconductor (see, e.g., Refs. 1 and 2). For such a superconductor,  $H_{c1}$  and a quantity assuming the role of  $H_{c2}$ , which we shall designate by  $H_g$ , are determined by the parameters of the Josephson-current density  $j_j$  and by the granule dimensions  $a$ :  $H_{c1} \sim j_j a$ ,  $H_2 \sim \Phi_0/a^2$  ( $\Phi_0$  is the fluxon). Experiment usually yields  $H_{c1} \sim 10^{-3}$  Oe (Ref. 3) and  $H_g \sim 10$  Oe (Ref. 4). The effective penetration depth  $\lambda^2 \sim \Phi_0/H_{c1}$  is large in this case. Since the role of the coherence length is played in this superconductor by the granule dimension  $a$ , the Landau–Ginzburg parameter is  $\kappa \sim \lambda/a \gg 1$ . In the field interval  $H_{c1} < H < H_g$  such a medium contains a macroscopic system of vortices with characteristic distances  $\delta \sim (\Phi_0/H)^{1/2} \gg a$  between them. It is obvious that the concept of an effective medium is applicable in this situation, since the vortices entrain many granules.

All of the above is basic equilibrium electrodynamics. Pinning, however, prevents equilibrium in ceramic HTSC, and the magnetic-field penetration must be described in this case in the language of nonequilibrium electrodynamics. It is customary then to use the premises of the theory of the critical state.

## THEORY

In critical-state theory the magnetic-pressure force  $\nabla p$  is balanced by the pinning force  $\alpha(h)$ . The corresponding equation for a low-field superconductor having an effective diamagnetic permeability  $\mu_{\text{eff}}$  (more below) is

$$|\nabla p| = \alpha(h), \quad p = \mu_{\text{eff}} h^2(x) / 8\pi, \quad (1)$$

where  $h(x)$  is the inhomogeneous magnetic field in the sample.

Equation (1) contains the absolute value  $|\nabla p|$  because

pinning is a dry-friction force and is therefore always directed counter to the magnetic-pressure force. Such a system is non-Hamiltonian, and hence the disequilibrium. Consider a slab of thickness  $d$  in the  $x$  direction and infinite in the two other directions. We direct the field along  $z$ . The critical-state equation (1) is then

$$\left| \frac{dh}{dx} \right| = 4\pi j_c(h), \quad j_c(h) = \frac{\alpha(h)}{\mu_{\text{eff}} |h|}. \quad (2)$$

The quantity  $j_c(h)$  proportional to the pinning force is called the critical-current density. Using the Maxwell equation  $4\pi j(x) = -dh/dx$ , where the current density  $j(x)$  is directed along  $y$ , we obtain  $|j(x)| = j_c(h)$ . Thus, the absolute value of the current density is  $j_c(h)$  at each point of space.

As seen from (2), the crucial quantity in critical-state theory is the phenomenological function  $j_c(h)$ , usually defined in a specific form, e.g.,

$$\begin{aligned} j_c(h) = j_0 \quad (i), \quad j_c(h) = j_0 \frac{H_0}{H_0 + |h|} \quad (ii), \\ j_c(h) = j_0 \frac{H_0}{|h|} \quad (iii), \quad j_c(h) = j_0 \frac{H_0^2}{H_0^2 + h^2} \quad (iv), \end{aligned} \quad (3)$$

where  $H_0$  is some characteristic field.

The relation (i) above leads to Bean's model,<sup>5,6</sup> model (ii) was proposed in Ref. 7, and (iii) is called the Kim–Anderson model.<sup>8</sup> There is in general no theoretically justified choice of the function  $j_c(h)$ . It is therefore obviously important to determine it experimentally. A  $j_c(h)$  dependence corresponding to the case (iv) is derived here by two independent methods.

Recall that in our situation the field hardly penetrates into the granules. This is accounted for by introducing<sup>1,2</sup> the concept of effective diamagnetic permeability  $\mu_{\text{eff}} = f_n + f_s \langle \mu_{\text{gr}} \rangle$ , where  $f_s$  and  $f_n$  are the fractions of the superconducting and nonsuperconducting media,  $f_n + f_s = 1$ , and  $\langle \mu_{\text{gr}} \rangle$  is the average granule permeability and depends on the London penetration depth, size, and shape of the granules. The induction in the sample can be represented in this case by

$$B(x) = \mu_{eff} h(x). \quad (4)$$

The penetration of the field  $h(x)$  is screened by the Josephson currents, which in conjunction with the pinning force actually determine  $j_c(h)$ . The critical-state equation has therefore been written just for  $h(x)$ . Expression (4) in fact constitutes an extension of  $h(x)$  to include the entire sample volume, recognizing that the field does not penetrate into the granules (see Refs. 1 and 2 for details).

The most important property of the critical state is impenetrability. Its mathematical manifestation is that Eq. (2) contains not the current density but only its absolute value. This leads (see Refs. 1, 2, 5, 6) to kinks on the field and induction distributions  $h(x)$  and  $B(x)$ , and also to hysteresis of these quantities and of their mean values  $b$  and  $\bar{B}$  over the sample:

$$b = \frac{1}{d} \int_{-d/2}^{d/2} h(x) dx, \quad \bar{B} = \mu_{eff} b. \quad (5)$$

This hysteresis was considered in detail, for example, in Ref. 2, and we shall use those results hereafter.

We apply collinearly to the sample a static field  $H$  and an alternating field of amplitude  $h_0$ ,  $H(t) = H + h_0 \cos(\omega t)$  and expand the induction  $B(t)$  in a Fourier series

$$\bar{B}(t) = -\frac{a_0}{2} + \sum_n [a_n \cos(n\omega t) + b_n \sin(n\omega t)], \quad (6)$$

$n=1, 2, 3, \dots$

The amplitudes of the harmonics  $a_n$  and  $b_n$  depend on the relation between the four fields  $h_0$ ,  $H$ ,  $H_0$ , and  $h_2$ , of which two ( $h_0$  and  $H$ ) are external, while  $H_0$  and  $h_2$  are determined by the properties of the superconductor itself and depend on temperature. The field

$$h_2 = 2\pi d j_0 \quad (7)$$

is governed also by the sample size.

Although a situation with arbitrary relations between these fields can be treated theoretically, we consider only two simple asymptotic cases that facilitate the comparison of the theory with experiment, and can be experimentally realized.

Low-field phenomena in HTSC are usually described by some arbitrary phenomenological function  $f_c(H)$  whose parameters are next determined from experiment. This approach is integral. Here, however, we use a differential approach and find two limiting cases in which the measured quantities are determined by the values of  $j_c(H)$ , where  $H$  is the applied static field. This enabled us to find the explicit form of  $j_c(H)$  directly from experiment.

We designate as the low-field regime the case  $h_0 \ll H$ ,  $H_0$ ,  $h_2$ , with arbitrary relations between  $H$ ,  $H_0$  and  $h_2$ .

The second limiting case is defined by the conditions  $h_0 \sim h_2(H)$  and  $h_2(H) \ll H_0$ , where  $h_2(H) = 2\pi d j_c(H)$  ( $h_2(0) \equiv h_2$ ). It is readily seen that the inequality  $h_2(H) \ll H_0$ , is equivalent to the condition

$$d \ll l = H_0 / 4\pi j_c(H), \quad (8)$$

where  $l$  is a certain characteristic length. The sample is called thin for  $d \ll l$  and thick for  $d \gg l$ . In a thin sample the

magnetic-field gradient can be regarded as independent of  $x$ , for when the condition (8) is satisfied the field variation over the sample dimension  $d$  is always less than  $H$  or  $H_c$ . It is thus always possible to expand  $j_c(H(t))$  in terms of the alternating field amplitude  $h_0$  and retain only the zeroth term of the expansion. The expression for the partial hysteresis loop [see Eq. (12) below] will be quite similar to the corresponding expression in Bean's model if  $h^*$  of Ref. 6 is replaced by  $h_2(H)$ .

Note that the weak-field concept holds for both thick and thin samples. We shall show below that the characteristic field  $H_0$  for the investigated sample is approximately 2 Oe and varies little with temperature in the interval from 78 to 90 K. In the same temperature interval, however,  $j_0$  varies over almost four orders of magnitude (from  $10^2$  to  $10^{-2}$  A/cm<sup>2</sup>). Our sample can therefore be regarded as thick at low temperature, when the current density  $j_c(H)$  is high, and as thin near  $T_c$ , when  $j_c(H) \rightarrow 0$ .

Consider the weak-field case:  $h_0 \ll h_2$ ,  $H_0$ ,  $H$ . According to Ref. 2, the expressions for the descending and ascending parts of the hysteresis loop are

$$\begin{aligned} \bar{B}_{asc}^{desc}(h) &= \mu_{eff} b_{asc}^{desc}(h), \quad b_{asc}^{desc}(h) = b_0/2 + \Delta b_{asc}^{desc}(h), \\ \Delta b_{asc}^{desc}(h) &= \frac{1}{4\pi d j_c(H)} [hh_0 \pm 1/2(h_0^2 - h^2)] \\ &+ \frac{1}{16\pi d} \frac{d}{dH} \left[ \frac{1}{j_c(H)} \right] [h_0 h^2 \pm h(h_0^2 - h^2)], \end{aligned} \quad (9)$$

$$h \equiv h(t) = h_0 \cos \omega t,$$

where  $b_0 = a_0/\mu_{eff}$  is independent of  $t$ . Expanding  $B(t)$  in a Fourier series, we obtain expressions for the harmonics<sup>2,6</sup>

$$\begin{aligned} a_1 &= \frac{\mu_{eff} h_0^2}{4\pi j_c(H) d}, \quad a_{2k+1} = 0, \quad k \geq 1, \\ b_{2k+1} &= -\frac{\mu_{eff} h_0^2}{8\pi^2 j_c(H) d} \frac{1}{(k^2 - 1/4)(k + 3/2)}, \quad k = 0, 1, 2, \dots, \\ a_2 &= \frac{\mu_{eff} h_0^3}{32\pi d} \frac{d}{dH} \left\{ \frac{1}{j_c(H)} \right\}, \quad a_{2k} = 0, \quad k \geq 2, \\ b_{2k} &= -\frac{\mu_{eff} h_0^3}{16\pi^2 d} \frac{d}{dH} \left\{ \frac{1}{j_c(H)} \right\} \frac{k}{(k^2 - 1/4)(k^2 - 9/4)}, \quad k = 1, 2, \dots \end{aligned} \quad (10)$$

Note that the even harmonics of a weak field are small, since the even part of the hysteresis loop in (9) is small, proportional to the parameter  $h_0/H_0$  for  $h < h_0$ , or to  $h_0/H$  for  $H_0 < H$ .

We consider now the limiting case of a thin sample, when  $h_0$  is of the same order as  $h_2$ , but  $h_0, h_2 \ll H_0$ . Here, as above, the even part of the hysteresis loop is small in proportion to the very same parameters. We write down only the odd part of the hysteresis loop that depends on the relation between  $h_0$  and  $h_2(H)$ .

For  $h_0 < h_2(H)$  we have for the hysteresis loop

$$\Delta b_{asc}^{desc}(h) = \frac{2hh_0 \pm (h_0^2 - h^2)}{4h_2(H)}. \quad (11)$$

For  $h_0 > h_2(H)$  the form of the hysteresis loop becomes dependent on the range of  $h$ :

$$\begin{aligned}
b_{\text{desc}}(h) &= h_0 - \frac{h_2(H)}{2} - \frac{(h_0-h)^2}{4h_2(H)}, & h_0 - 2h_2(H) < h < h_0, \\
b_{\text{desc}}(h) &= h + \frac{h_2(H)}{2}, & -h_0 < h < h_0 - 2h_2(H), \\
b_{\text{asc}}(h) &= -h_0 + \frac{h_2(H)}{2} + \frac{(h_0+h)^2}{4h_2(H)}, & \\
& -h_0 < h < -h_0 + 2h_2(H), \\
b_{\text{asc}}(h) &= h - \frac{h_2(H)}{2}, & -h_0 + 2h_2(H) < h < h_0.
\end{aligned} \tag{12}$$

Using the expansion (6) and Eqs. (11) and (12) we obtain for the harmonics expressions corresponding to three different relations between  $h_0$  and  $h_2(H)$ : a)  $h_0 < h_2(H)$ ; b)  $h_2(H) < h_0 < 2h_2(H)$  and c)  $h_0 > 2h_2(H)$ .

In the first of these cases the expressions for  $a_1$  and  $b_{2k+1}$ , viz.,

$$\begin{aligned}
a_1 &= \frac{\mu_{eff} h_0^2}{2h_2(H)}, & a_{2k+1} &= 0, & k &\geq 1, \\
b_{2k+1} &= -\frac{\mu_{eff} h_0^2}{4\pi h_2(H)} \frac{1}{(k^2 - 1/4)(k + 1/2)},
\end{aligned} \tag{13}$$

coincide with the corresponding expressions for a weak field [see (10)], if it is recognized that  $h_2(H) = 2\pi dj_c(H)$ . Note, however, that expressions (13), in contrast to (1), are valid not only for  $h_0 \ll h_2(H)$  but also for all  $h_0 \leq h_2(H)$ .

In the second case  $h_2(H) < h_0 < 2h_2(H)$  we obtain

$$\begin{aligned}
a_{2k+1} &= \frac{\mu_{eff} h_0^2}{2h_2(H)} \delta_{k,0} + \frac{\mu_{eff} h_0^2}{8\pi h_2(H)} \frac{\sin[(2k+1)x_2]}{k(k+1)(2k+1)} - \frac{\mu_{eff} h_0^2}{4\pi h_2(H)} \\
& \times \frac{\cos(2x_2) \sin[(2k+1)x_2]}{(k+1)(2k+1)(2k+3)} \\
& - \frac{3\mu_{eff} h_0^2 \sin[(2k-1)x_2]}{8\pi h_2(H) k(k+1)(2k-1)(2k+3)}, \\
b_{2k+1} &= -\frac{\mu_{eff} h_0}{2\pi} \frac{1}{k(k+1)(2k+1)} \\
& - \frac{3\mu_{eff} h_0^2}{4\pi h_2(H) k(k+1)(2k-1)(2k+1)(2k+3)} \\
& + \frac{\mu_{eff} h_0^2}{8\pi h_2(H)} \frac{\cos[(2k+1)x_2]}{k(k+1)(2k+1)} \\
& - \frac{\mu_{eff} h_0^2}{4\pi h_2(H)} \frac{\cos(2x_2) \cos[(2k+1)x_2]}{(k+1)(2k+1)(2k+3)} \\
& - \frac{3\mu_{eff} h_0^2}{8\pi h_2(H)} \frac{\cos[(2k-1)x_2]}{k(k+1)(2k-1)(2k+3)},
\end{aligned} \tag{14}$$

where

$$\cos x_2 = \frac{2h_2(H)}{h_0} - 1.$$

In the last case  $h_0 > 2h_2(H)$  we have finally

$$\begin{aligned}
a_{2k+1} &= \mu_{eff} h_0 \delta_{k,0} + \frac{\mu_{eff} h_0^2}{8\pi h_2(H)} \frac{\sin[(2k+1)x_1]}{k(k+1)(2k+1)} \\
& - \frac{\mu_{eff} h_0^2}{4\pi h_2(H)} \frac{\cos(2x_1) \sin[(2k+1)x_1]}{(k+1)(2k+1)(2k+3)} \\
& - \frac{3\mu_{eff} h_0^2}{8\pi h_2(H)} \frac{\sin[(2k-1)x_1]}{k(k+1)(2k-1)(2k+3)}, \\
b_{2k+1} &= -\frac{\mu_{eff} h_0}{2\pi} \frac{1}{k(k+1)(2k+1)} \\
& - \frac{3\mu_{eff} h_0^2}{4\pi h_2(H)} \frac{1}{k(k+1)(2k-1)(2k+1)(2k+3)} \\
& - \frac{\mu_{eff} h_0^2}{8\pi h_2(H)} \frac{\cos[(2k+1)x_1]}{k(k+1)(2k+1)} \\
& + \frac{\mu_{eff} h_0^2}{4\pi h_2(H)} \frac{\cos(2x_1) \cos[(2k+1)x_1]}{(k+1)(2k+1)(2k+3)} \\
& + \frac{3\mu_{eff} h_0^2}{8\pi h_2(H)} \frac{\cos[(2k-1)x_1]}{k(k+1)(2k-1)(2k+3)},
\end{aligned} \tag{15}$$

where

$$\cos x_1 = 1 - \frac{2h_2(H)}{h_0}.$$

It follows from (13)–(15) that  $h_0$  varies in three intervals corresponding to different forms of the hysteresis loop with different expressions for the harmonics in these regions. One might conclude at first glance that the expressions for  $a_{2k+1}$  and  $b_{2k+1}$  have singularities at  $h_0 = h_2(H)$  and  $h_0 = 2h_2(H)$  respectively. It is clear from physical considerations, however, that only  $h_0 = h_2(H)$  can be a singular point, and not  $h_0 = 2h_2(H)$ . In fact, analysis shows that (14) and (15) have no singularities at all at  $h_0 = 2h_2(H)$ , and the cause of the two different expressions for  $a_{2k+1}$  and  $b_{2k+1}$  is that  $\cos x_2$  must be replaced by  $\cos x_1$  when  $x_2$  goes through  $\pi/2$ .

It is clear next from (14) and (15) that the arguments of the cosines and the sines ( $x_2$ , and later also  $x_1$ ) vary from 0 to  $\pi/2$  and back. Functions such as  $\cos[(2k+1)x_1]$  also oscillate and the number of their oscillations increases with  $k$ . It is clear from (14) and (15) that these oscillations are due to the kink on the hysteresis loop in (12) at  $|h| = h_0 - 2h_2(H)$ . Such a kink is in fact a threshold singularity and is the cause of the oscillations. We shall show below that the oscillations in our case took the form of one maximum in the dependence of the magnitude of the third-harmonic amplitude on  $h_0$ . Note that oscillations of even harmonics as functions of the static field were observed in Ref. 9.

It follows from (13)–(15) that the susceptibilities  $a_{2k+1}/h_0$  and  $b_{2k+1}/h_0$ , for which

$$\begin{aligned}
\frac{a_1}{h_0} &= 1 + 4\pi\chi_1'(y), & \frac{a_{2k+1}}{h_0} &= 4\pi\chi_{2k+1}'(y) \quad (k \geq 1), \\
\frac{b_{2k+1}}{h_0} &= 4\pi\chi_{2k+1}''(y),
\end{aligned} \tag{16}$$

are functions of the dimensionless variable  $y = h_0/h_2(H)$ . The equations in (16) are quite remarkable. Indeed, if  $h_0, h_2 \ll H_0$ , only, the entire dependence of the susceptibilities on temperature and field is contained in the scaling variable  $y$ , and their dependence on  $y$  is itself universal. We present by way of example, using (13)–(16), the dependence of  $\chi''_1, \chi''_3$  and  $\chi'_e$  on  $y$ ;

$$\begin{aligned}
 & \text{For } y < 1 \\
 4\pi\chi''_1 &= \frac{2}{3\pi} \mu_{eff} y, \quad 4\pi\chi'_3 = 0, \quad 4\pi\chi''_3 = -\frac{2}{15\pi} \mu_{eff} y. \\
 & \text{For } y > 1 \\
 4\pi\chi''_1 &= \frac{2}{\pi} \mu_{eff} \left[ \frac{1}{y} - \frac{2}{3y^2} \right], \quad 4\pi\chi'_3 = -\frac{32}{15\pi} \mu_{eff} \frac{(y-1)^{3/2}}{y^4}, \\
 4\pi\chi''_3 &= -\frac{2}{15\pi y} \mu_{eff} \left[ \frac{16}{y^3} - \frac{40}{y^2} + \frac{30}{y} - 5 \right]. \quad (17)
 \end{aligned}$$

It can be seen from (17) that  $\chi'_{2k+1}$  and  $\chi''_{2k+1}$  are indeed continuous at  $y = 2$  ( $h_0 = 2h_2(H)$ ) and that they are oscillatory functions of  $y$  since, for example, the polynomial in  $\chi''_3$  has two extrema. This behavior is shown in Fig. 1. It is noteworthy that oscillations of  $\chi''_3$  with temperature were observed in Ref. 10.

## EXPERIMENT

The principal task of the experimental part of our work was to study the static and dynamic magnetic characteristics of an ceramic HTSC and compare the results with the theory. We have already mentioned that the penetration of a magnetic field into a ceramic superconductor is determined to a considerable degree by the relations between the fields  $h_0, H, h_2$ , and  $H_0$ . Wide ranges of temperatures and external fields were used to achieve various situations in experiment, including the asymptotic “weak field” and “thin sample” cases.

The investigations have established that the above theory describes the observed phenomena adequately. It was therefore possible to determine a number of important pa-

rameters indicative of magnetic-field penetration into a superconductor, including the critical field  $j_0$  and its temperature dependence, the specific form of the function  $j_c(H)$ , and the dependence of  $\mu_{eff}$  on  $T$ . The  $j_c(H)$  dependence in the limiting cases of a weak field and a thin sample were found to have the same form, corresponding to (iv) in Eq. (3). It was also possible to determine the temperature dependence of the characteristic fields  $h_2(T)$  and  $H_0(T)$ .

The ceramic investigated was  $Y_1Ba_2Cu_3O_7$  ( $\rho \approx 5$  g/cm<sup>3</sup>) produced by a procedure described in Ref. 11. The sample shapes varied with the purpose of the experiment.

A cylindrical sample ( $\approx 1$  mm dia,  $l \approx 7$  mm) was used to study the temperature dependence of the static susceptibility  $\chi_{dc} = M/H$  ( $M$  is the magnetization). A spherical sample ( $\approx 1.2$  mm dia) was used to determine  $\mu_{eff}$ . A non-standard SQUID magnetometer<sup>12</sup> in conjunction with a UJ 111 pickup and a gradiometer of the first kind were used to measure these two samples. All the data obtained by this method take into account the demagnetization factor. A toroidal sample with outside and inside diameters 12.5 and 7.5 mm were used to study the field and temperature dependence of the real and imaginary susceptibility ( $\chi'$  and  $\chi''$ ) as well as of the magnitudes of the higher-order-harmonic amplitude  $c_n = (a_n^2 + b_n^2)^{1/2}$ . A single-layer toroidal measuring coil was wound on the sample. Obviously, the toroidal shape of the sample excludes demagnetization effects and permits a more correct comparison of the experimental and theoretical results. The values of  $\chi'$  and  $\chi''$  were measured at 20 and 100 kHz, and the higher harmonics at the fundamental frequency 20 kHz. These experiments (the measurement procedure is described in Ref. 13) were performed in fields  $10^{-2} < h_0, H < 2.5$  Oe. The ambient laboratory field was less than 0.01 Oe in all experiments.

We begin the survey of the experiments with the temperature dependence of the susceptibilities. The most distinctive feature of the temperature dependence of the dynamic quantities is the presence of one or two maxima on the  $c_3(T)/h_0$  dependence (Fig. 2). The high-temperature maximum is observed only at relatively high alternating-field amplitudes. It can be seen from the figure that the location of the low-temperature maximum depends strongly on the field amplitude. Note that the plots of  $c_5(T)/h_0$  and

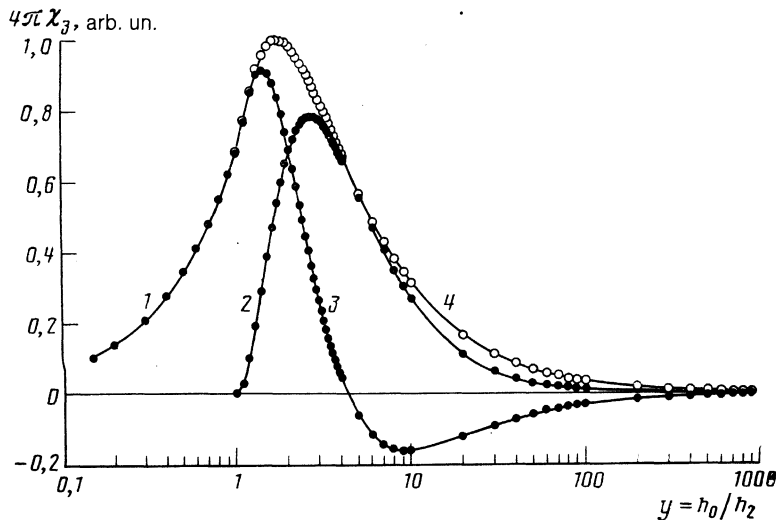


FIG. 1. Dependence of the third harmonic of the susceptibility on  $y = h_0/h_2(H)$  as calculated from Eq. (7):  $-4\pi\chi''_3$  (curve 1 for  $y < 1$  and curve 3 for  $y > 1$ );  $-4\pi\chi'_3$  (curve 2);  $|4\pi\chi_3| = 4\pi[(\chi'_3)^2 + (\chi''_3)^2]^{1/2}$  (curves 1 and 4 for  $y < 1$  and  $y > 1$ , respectively).

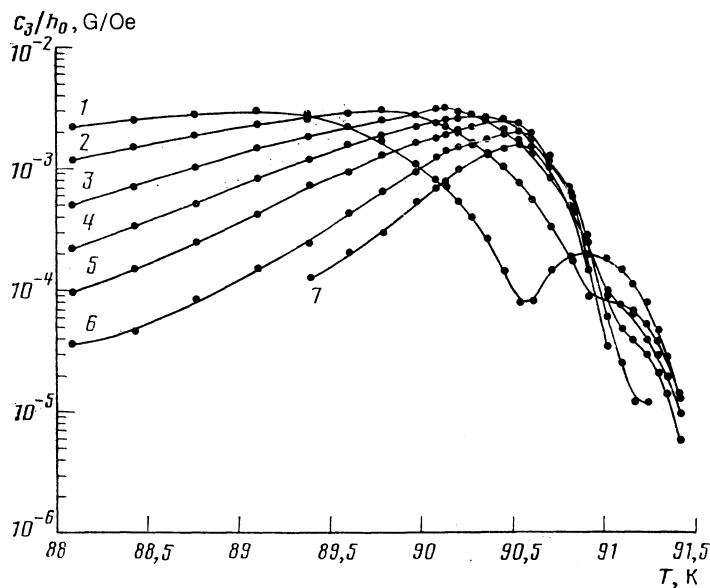


FIG. 2. Temperature dependence of  $c_3(T)/h_0$  (1), obtained in the absence of an external dc static magnetic field at various amplitudes of the alternating field  $h_0 = 1$  (1), 0.5 (2), 0.2 (3), 0.1 (4), 0.05 (5), 0.02 (6), 0.01 (7) Oe.

$b_1(T)/h_n = 4\pi\chi''(T)$  also have two maxima each. A similar form of  $\chi''(T)$  was observed, for example, in Refs. 14 and 15 and in a number of other studies. This behavior of the harmonics arises because the present pattern of ultraweak-field penetration into a ceramic superconductor is indeed complicated by the field penetration into the granules themselves near  $T$ . It is believed at present (see, e.g., Refs. 14 and 15) that the high-temperature maximum is due to field penetration into the granules, and that the low-temperature maximum is due to field penetration only into the weakly bonded intergranular medium.

The presence of two (intragranular and intergranular) different mechanisms of field penetration into HTSC is manifested in the temperature dependence of the static susceptibility. The  $\chi_{dc}(T)$  dependence (Fig. 3) clearly shows a bend at  $T \approx 89$  K that separates two temperature regions of the susceptibility variation corresponding to maxima at  $c_3(T)/h_0$ . No detailed study, however, was made here of the field penetration into the granules (of the high-temperature maximum).

Let us examine now the results for the weak field ( $h_0 \ll H, H_0, h_2$ ) regime. In this limiting case, according to

(10), the odd harmonics should increase with the field  $h_0$  as  $h_0^2$  and the even (at  $H = 0$ ) as  $h_0^3$ . The harmonics  $a_1, b_1, c_3, c_5$  and  $c_2$  had this dependence on  $h_0$  in the temperature interval from 78 to 89 K, both for  $H = 0$  and for finite values of the static field (Fig. 4). Naturally, no even harmonics were observed at  $H = 0$ .

The coefficients of  $h_0^2$  directly related to the critical current  $j_0$  were determined from the dependence of  $a_1, b_1, c_3$ , and  $c_5$  on  $h_0$ . The temperature dependence of these coefficients is shown in Fig. 5. We stress that although the harmonics themselves change by two orders of magnitude in the temperature range, the ratios  $a_1/b_1, b_1/c_3$  etc. are practically independent of temperature (see the plot of  $a_1/b_1$  in Fig. 5). This confirms once more that these experiments were performed under weak-field conditions. Indeed, all the expressions for  $a_1$  and  $b_{2k+1}$  in (10) contain the ratio  $\mu_{eff}(T)/j_c(H, T)$ . It is obvious then that  $a_1/b_1, b_1/c_3$ , etc. need not depend on  $\mu_{eff}(T)$  and  $j_c(H, T)$ , and hence on the temperature. Note that these harmonic ratios are close to those calculated from (10) and practically coincide with the data of Ref. 16.

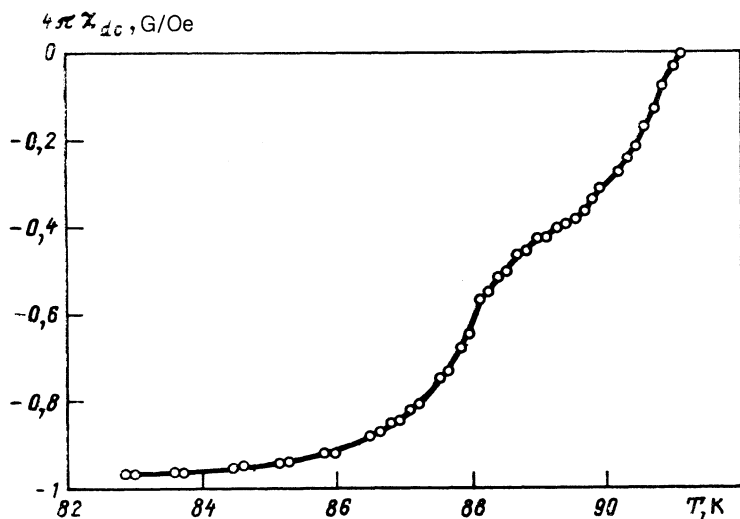


FIG. 3. Temperature dependence of  $4\pi\chi_{dc}$  obtained for cooling in a zero external field followed by turning on a measuring field  $H = 0.4$  Oe (cylinder).

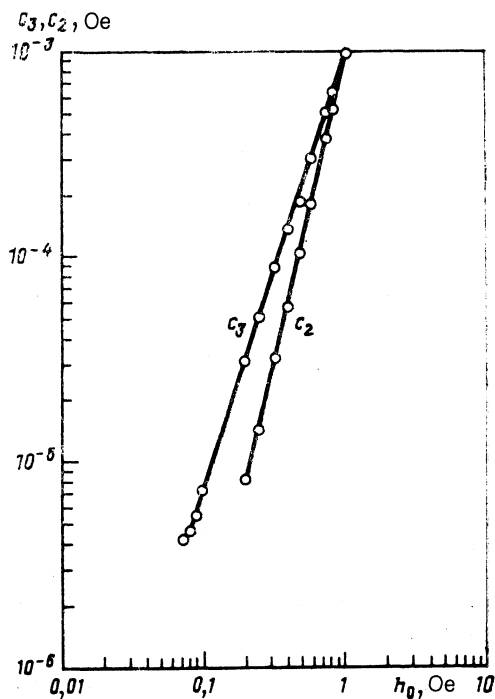


FIG. 4. Plots of  $c_3(h_0)$  and  $c_2(h_0)$  at  $H = 1.5$  Oe and  $T = 86$  K.

In the weak-field regime the harmonic amplitudes, as noted, are directly connected with the critical-current density  $j_0$ . It is impossible, however, to calculate  $j_0$  only from  $a_1$  and  $b_{2k+1}$ , since knowledge of  $\mu_{\text{eff}}$  is also necessary. It was necessary therefore to determine the effective diamagnetic permeability independently. This was done by determining  $\mu_{\text{eff}}$  from the high-field asymptote of the hysteresis loop or from the magnetization curve obtained in static field  $H$ . Note that "high-field asymptote" means the field region  $H \gg H_0, h_2$ . In this case  $j_c(H) \rightarrow 0$  there is no screening due to pinning, and the induction  $\bar{B}$  is simply  $\bar{B} = \bar{B}_{\text{desc}} = \bar{B}_{\text{asc}} = \mu_{\text{eff}} h$  (see Ref. 2 for details). A plot of  $\mu_{\text{eff}}(T)$

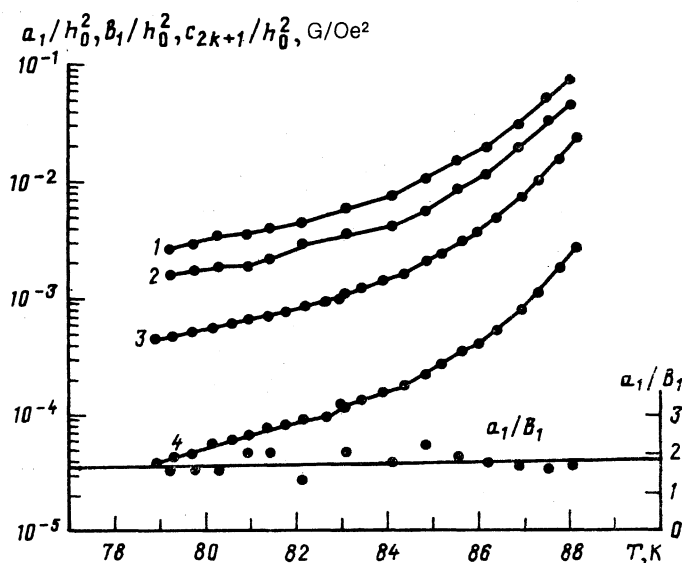


FIG. 5. Plots of  $a_1/h_0^2$  (1),  $b_1/h_0^2$  (2),  $c_3/h_0^2$  (3),  $c_5/h_0^2$  (4) and  $a_1/b_1$ , obtained in a zero external stationary field.

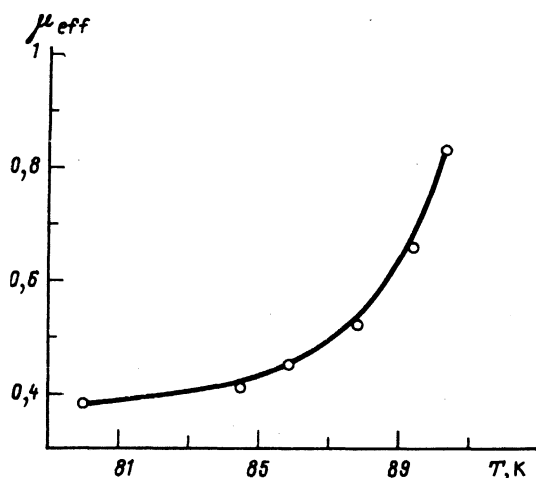


FIG. 6. Temperature dependence of the effective diamagnetic permeability  $\mu_{\text{eff}}$  determined from intensity measurements.

obtained in this manner is shown in Fig. 6. Clearly, the effective permeability changes most strongly near  $T_c$ .

The principal quantity in the critical-state theory is, as already noted, the function  $j_c(H)$ . To determine this function explicitly we have used here a method proposed in Ref. 16. In this method, as seen from (10), the odd harmonics in weak fields are essentially proportional to  $1/j_c(H)$  and the even to  $(d/dH)(1/j_c(H))$ . Measurement of these harmonics as functions of the static fields at a fixed value of  $h_0$  therefore yields a self-consistent explicit form of  $j_c(H)$ . We performed such experiments with the third and second harmonics. To satisfy the weak-field conditions, the range of the alternating field  $h_0$  was chosen so that  $c_3$  and  $c_2$  depended quadratically and cubically on  $h_0$ , respectively.

As a result, when the static field varied from 0 to 1.5 Oe the third-harmonic amplitude, i.e.,  $1/j_c(H)$  increased like  $H^2$ , while the second-harmonic amplitude, i.e.,  $(d/dH)(1/j_c(H))$ , was linear in  $H$  (Fig. 7). Obviously, this behavior of the harmonics corresponds to

$$j_c(H) = j_0 \frac{H_0^2}{H_0^2 + H^2} \quad (18)$$

which is consistent with the model (iv) of Eq. (3).

In this case we have  $c_3(H)/c_3(0) - 1 = H^2/H_0^2$ . Plots of  $c_3(H)$  measured at various temperatures show that the characteristic field  $H_0$  ranges from 3 to 1.8 Oe in the interval  $78 < T < 86$  K (see Fig. 12 below). We know of no observation of a  $j_c(H)$  dependence in the form (18) in classical superconductors or in ceramic HTSC. It appears that the only attempt to determine the explicit form of  $j_c(H)$  in low-field electrodynamics is reported in Ref. 16. However, the  $j_c(H)$  dependence obtained there for another ceramic, Y-Ba-Cu-O, is of the form (ii), i.e.,  $j_c(H) = j_0 H_0 / (|H| + H_0)$ . The causes of the different forms of  $j_c(H)$  are still unclear. Note that a dependence of the form (ii) was observed earlier<sup>7</sup> in classical superconductors in the high-field region.

The aim of the succeeding experiments was to determine the behavior of the susceptibility in a sample that can be regarded as thin, i.e., under the condition  $d \ll l = H_0 / 4\pi j_c(H)$ . In this case, as already mentioned, the theory predicts that all the susceptibilities ( $\chi'$ ,  $\chi''$ ,  $\chi_3$ , etc.)

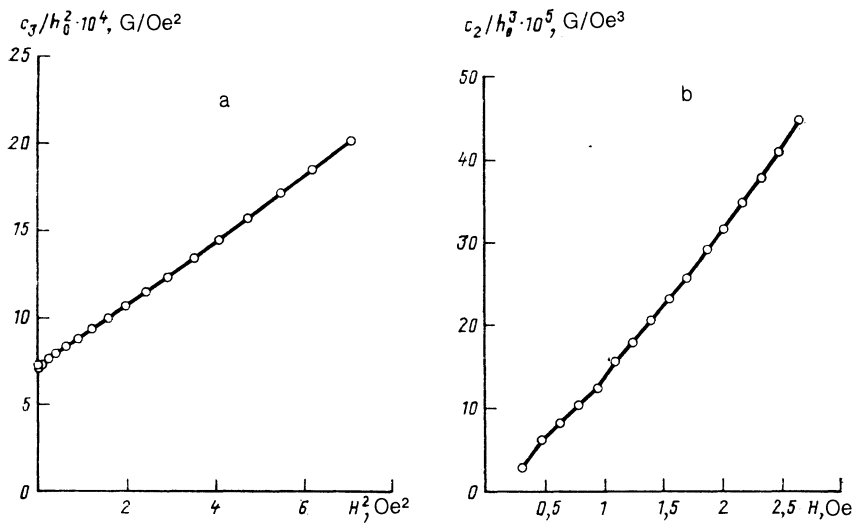


FIG. 7. a) Dependence of  $c_3/h_0^2$  on  $H^2$ , b) dependence of  $c_2/h_0^3$  on  $H$ ;  $T = 83.92$  K, ( $h_0 = 0.25$  Oe).

scale with the dimensionless variable  $y = h_0/h_c$ . Obviously, this regime can be observed in this geometry when the current density  $j_0$  is low, or else by decreasing  $j_c(H)$  using a strong magnetic field  $H \gg H_0$ . It was impossible, however, to obtain in our experiments a field of sufficient strength to produce the "thin-sample" state. This regime could therefore be observed only for small  $j_0$ , i.e., near  $T_c$ .

It follows from (17) that for  $h_0 \approx h_2 \ll H_0$  the function  $4\pi|\chi_3(y)|$  should have a maximum (see Fig. 1). This maximum, which is typical of the thin-sample regime, is clearly seen on the  $c_3(T)/h_0$  plot of Fig. 2 (a change of temperature is accompanied by one of  $h_2$  and hence of  $y$ ). To determine the scaling behavior of the susceptibility in this limiting case, we studied the magnitude of the third harmonic  $c_3$  as a function of the alternating-field amplitude  $h_0$  for various values of the static field and of the temperature. Typical plots of  $c_3(h_0)/h_0 = 4\pi|\chi_3(h_0)|$  for different  $H$  (at constant  $T$ ) are shown in Fig. 8. Clearly, the plots have maxima at a certain field amplitude  $h_0 = h_{\max}$  with  $h_{\max}$  dependent on  $H$  (and, in general, on  $T$ ).

We introduce a new variable  $u = h_0/h_{\max}$  and replot Fig. 8 for this coordinate (Fig. 9). It is seen from the latter that all the  $c_3(u)/h_0$  dependence are practically equal. A

disparity is noticeable only for weak static fields. Susceptibility scaling is thus observed in strong static fields. In weak fields we have  $j_c(H) \approx j_0$  and the condition  $2\pi dj_0 = h_2 \ll H_0$  is not met. Indeed, extrapolation of the low-field values of  $H_0$  yields  $H_0 \approx 1.5$  Oe near  $T_c$ . On the other hand, it follows from the very same data for  $j_0$  that, for example,  $h_2 \approx 1.5$  Oe at  $T \approx 89.6$  K; i.e., the thin-sample criterion is not satisfied in this case as  $H \rightarrow 0$ .

The value of  $j_c(H)$  [and hence of  $h_2(H)$ ] decreases in strong static fields, and the sample can become "thin." As a check on this assumption, let us compare the  $c_3(h_0)/h_0$  plots (Fig. 10) obtained at different temperatures and for two values of the static field,  $H = 0$  (curves 1–3) and 1.95 Oe (curves 4–8). Evidently, in a strong field the plots of  $c_3(h_0)/h_0$  are in rather good agreement, but they differ for the same temperatures if  $H = 0$ . We regard this as convincing evidence favoring the existence of a thin-sample regime in which all the susceptibilities are universal functions of only one variable  $u = h_0/h_{\max}$ .

In the above comparison of the  $c_3(h_0)/h_0$  plots it was assumed implicitly that  $h_{\max} = \beta h_2$ , i.e.,  $y = \beta u$ , where  $\beta$  is a certain coefficient. It seems most correct to determine  $\beta$  from the  $c_3(h_0)/h_0$  dependence obtained in the region

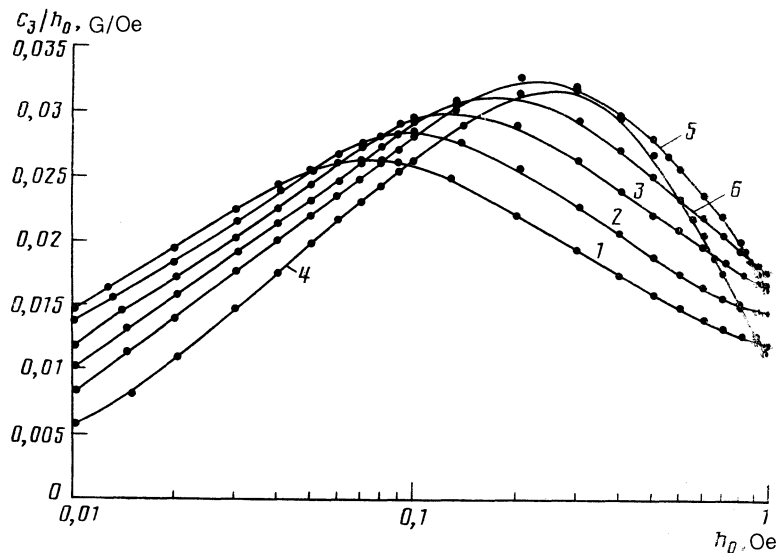


FIG. 8. Plots of  $c_3(h_0)/h_0$  vs  $h_0$  at  $H = 2.33$  (1), 1.95 (2), 1.55 (3), 1.17 (4), 0.78 (5), and 0.0 (6) Oe;  $T = 89.62$  K.

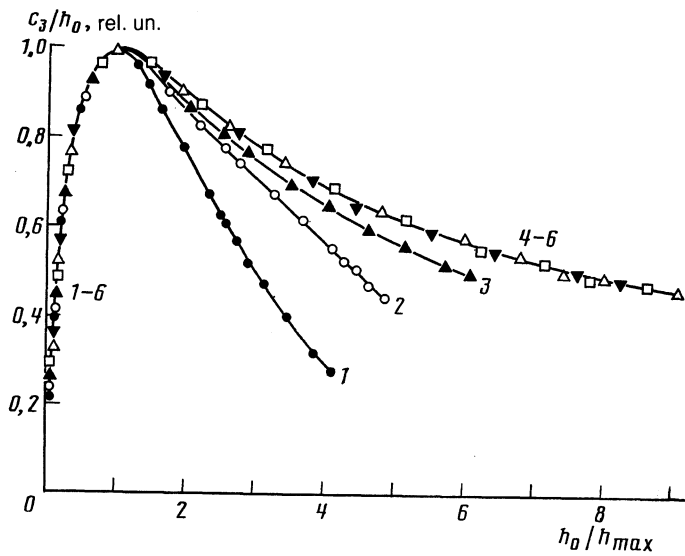


FIG. 9. Plots of  $c_3(h_0)/h_0$  vs  $u = h_0/h_{\max}$  at  $H = 0$  (1), 0.78 (2), 1.17 (3), 1.55 (4), 1.95 (5) and 2.33 (6) Oe;  $T = 89.62$  K.

where the two regimes overlap, i.e., when variation of the alternating-field amplitude in a sufficiently large interval simultaneously produces maxima on these plots as well as a weak-field regime ( $c_3 \sim h_0^2$ ). This situation actually existed, but only in a rather narrow temperature range ( $\leq 0.4$  K). In this case, by determining the coefficient of  $h_0^3$  from  $c_3(h_0)$  for  $h_0 \ll h_{\max}$  and then  $f_0$  by taking into account  $\mu_{\text{eff}}$  from (10), we can also calculate  $h_2 = 2\pi dj_0$ . On the other hand, putting  $h_{\max} = \beta h_2$  we obtained  $\beta = 1$ , i.e.,  $h_{\max}$  is none other than  $h_2$ .

According to the theory [see Eq. (17) and Fig. 1], however, a maximum of  $|4\pi\chi_3(y)|$  should be observed for  $h_0/h_2 \approx 1.7$  ( $\beta \approx 1.7$ ). Experiment, however, yields  $\beta = 1$ . This discrepancy can be attributed to the fact that the sample used in the experiments was toroidal, whereas expression (17) was derived for a slab. The following, however, must be noted. Since a universal dependence of the susceptibility on  $h_0/h_{\max}$  is observed in experiment, expression (17) can be rewritten as  $4\pi|\chi_3(u)| = \gamma f(u)$ , where  $f(1) = 1$ . The experimentally determined  $\gamma_{\text{exp}} \approx 0.034$  agrees quite well with its value  $\gamma_{\text{th}} = (2/15\pi)\mu_{\text{eff}} \approx 0.030$  calculated from Eq. (17).

Starting from the observed susceptibility scaling and assuming that  $h_{\max}(H) = h_2(H) = 2\pi dj_c(H)$ , we can determine from the dependence of  $h_{\max}$  on  $H$  (at constant  $T$ ) the explicit form of the function  $j_c(H)$  for temperatures at which the sample can be regarded as thin. As seen from Fig. 11,  $h_{\max}^{-1}$  is linear in  $H^2$ . Thus, for a thin sample  $j_c(H)$  has the same form as in the low-field regime, i.e.,  $j_c(H) = j_0 H_0^2 / (H^2 + H_0^2)$ . The approximate value of  $H_0$ , 1.3 Oe, determined in this case agrees quite well with a linear extrapolation of  $H_0(T)$ , in the weak-field regime, into this temperature region.

The critical current density  $j_0$ , as already mentioned, is one of the most important quantities in critical-state theory. We have determined  $j_0$  here in two limiting cases—weak field and thin sample. Recall that for the former case  $j_0$  was calculated with allowance for  $\mu_{\text{eff}}$ , using Eqs. (10), from the values of the coefficients of  $h_0^2$ , while in the latter it was calculated from the values of  $h_{\max}$  for an infinite cylindrical sample of diameter  $d = 0.25$  cm. In the present study we have thus determined the behavior of  $j_0$  in a wide temperature range.

For a more transparent representation of the relation

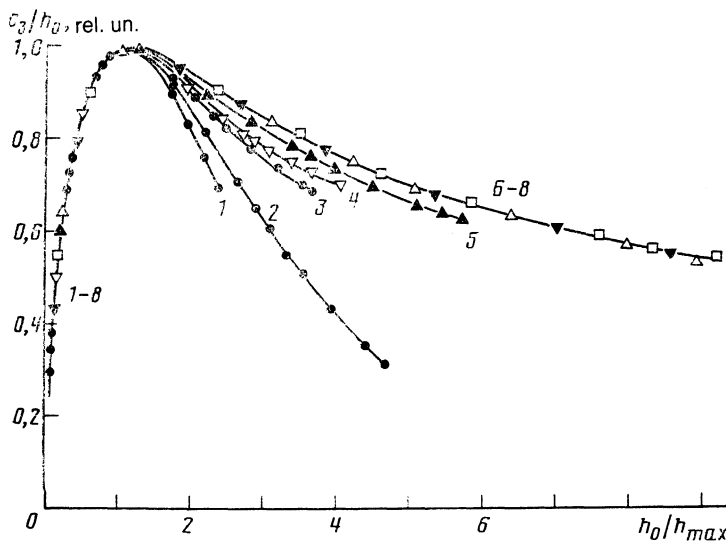


FIG. 10. Plots of  $c_3/h_0$  vs  $u = h_0/h_{\max}$ , obtained at various temperatures for two values of the static field:  $H = 0$  (curves 1-3) and 1.95 Oe (curves 4-8); curves 1-3 and 4-6 pertain to  $T = 89.07, 89.3, 89.62$  K, and curves 7 and 8 to  $T = 89.83$  and  $89.92$  K respectively.



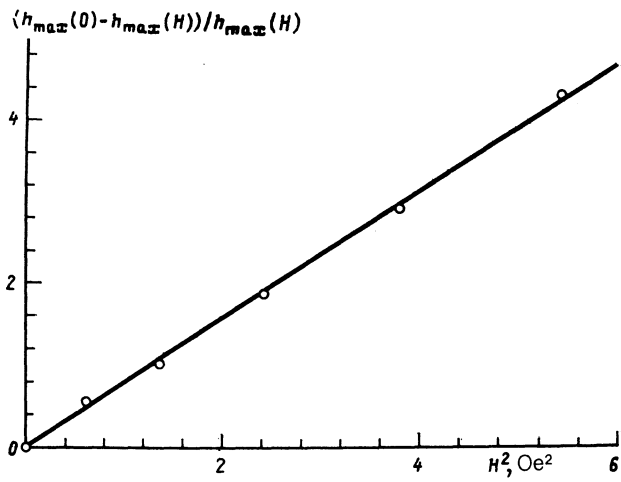


FIG. 11. Dependence of  $(h_{\max}(0) - h_{\max}(H))/h_{\max}(H)$  on  $H^2$ , where  $h_{\max}$  corresponds to  $H = 0$  and  $h_{\max}(H)$  to a finite value of the static field.

between the characteristic fields ( $h_0$ ,  $H$ ,  $H_2$ , and  $H_0$ ), and by the same token the conditions for realizing various limiting regimes, it is better to consider the function  $h_2(T)$  rather than  $j_0(T)$ . The functions  $h_2(T)$  and  $H_0(T)$  obtained in the two limiting cases are shown in Fig. 12. We see that, firstly, the characteristic field  $H_0$  depends little on temperature. Secondly,  $h_2$  is approximately equal to  $H_0$  at  $T \approx 89.6$  K, so that in the absence of an external dc field the thin-sample regime could be observed in our case only at higher temperatures. Thirdly, between 78 and 91 K the critical-current density  $j_0$  (whose numerical values can be obtained by multiplying  $h_2$  on this figure by  $1/2\pi d \approx 0.5 \text{ cm}^{-1}$ ) changes by almost four orders, the variation of  $j_0$  being strongest near  $T_c$ . This is indeed the cause of the small temperature interval in which the thin-sample case is observed. Note that for convenience Fig. 12 has two scales—for  $h_2$  and  $j_0$ .

Let us examine now the cause of this strong dependence of  $j_0$  (equivalently, of  $\alpha$ ) on  $\tau = (T_c - T)/T_c$ . We note first that even for an ordinary type-II superconductor  $f_0$  is the product of the pair-breaking current  $j_{c0}$  by the dimensionless quantity  $\zeta$  that describes directly the vortex-pinning force (see, e.g., Refs. 17 and 18). In our case the pair-breaking current is determined by the characteristic value of the Josephson bonds with  $j_{c0}$  far from the transition and  $j_{c0}$  near it, just as in an ordinary superconductor.<sup>19</sup> In view of the complex geometric structure of a Josephson medium, it is of course difficult to calculate  $\zeta(\tau)$ , but if the pinning is strong, then  $\zeta \ll 1$  and depends weakly on  $\tau$  (see the problem of strong pinning in a layered superconductor<sup>18</sup>). In this case  $j_c$  and  $\alpha$  are determined almost completely by the pair-breaking current and are proportional to  $\tau$  or  $\tau^{3/2}$ .

It can thus be concluded from our investigations that the critical-state theory can be used to describe the penetration of a magnetic field into granular superconductors. Various limiting cases were realized in experiments performed on HTSC in a wide range of temperatures and external fields, using different procedures, and important quantities indicative of the penetration of ultraweak fields in ceramic HTSC were determined.

We note in conclusion that low-field electrodynamics apparently also occurs in single crystals (see, e.g., Refs. 20–

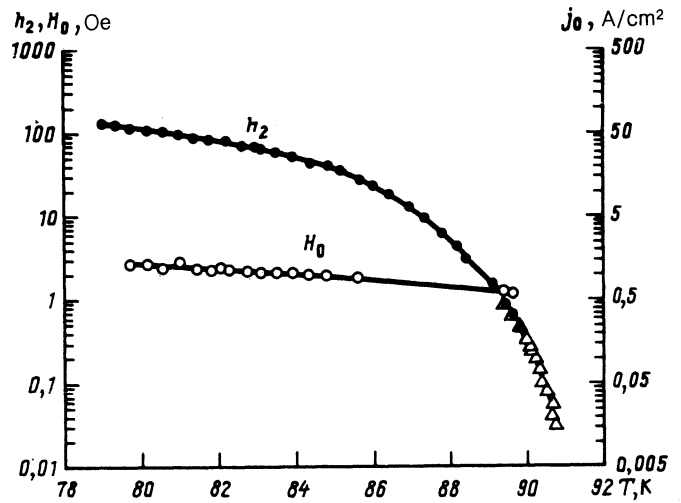


FIG. 12. Temperature dependence of the characteristic fields  $h_2 = 2\pi d j_0$  and  $H_0$  ( $\circ$ — $H_0$ ;  $\bullet$  and  $\Delta$  are the values of  $h_2$  determined for the case of a weak field and a thin sample, respectively). The values of  $H_0$  at  $T \approx 90$  K were determined from the  $h_{\max}(H)$  dependence. The scale of  $j_0$  is on the right.

22). In all likelihood, the role of weak bonds is assumed there by twins, and the role of granules by twinless regions. This entire group of phenomena is closely connected with the fact that the coherence length in HTSC systems is very small.<sup>23</sup> This very interesting question, however, is still open.

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