

# Type I superconductors in a high-amplitude, low-frequency alternating field

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We studied experimentally the destruction of superconductivity in long cylindrical samples of tin and indium by a magnetic field parallel to the axis of the cylinder and in the presence of an alternating electromagnetic field of large magnitude. Frequencies were employed at which the skin depth in the normal state was much less than the thickness of the sample. It is shown that as a result of the periodic process accompanied by the transitions on the surface, from the superconducting to the normal state, and vice versa, the magnetic induction flux is crowded into the interior of the sample. Because of this crowding, the transition of the entire sample to the normal state under the influence of the alternating field occurs in an external field  $H < H_c$ .

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The behavior of long cylinders of type-I superconductors, placed in a constant magnetic field  $H < H_c$  parallel to the axis of the cylinder, under the action of a low-frequency alternating field  $H_{\sim}(t) = H_{\sim} \cos \omega t$  ( $H + H_{\sim} > H_c$ ,  $\omega \leq 2\pi \times 10^4$ ) has been studied experimentally in Refs. 1 and 2. The aim of these experiments was to obtain information on the velocity of the boundary between the superconducting and normal phases and on the processes of formation of nuclei of a new phase. The motion of the interphase boundary in an alternating, low-frequency field has been studied theoretically in Ref. 3. It followed from calculation that destruction of superconductivity in the external field  $H < H_c$  can take place only at distances of the order of the depth of the skin layer  $\delta_{sk}$ . The fact that the interior of the metal remains in the superconducting state seemed so obvious that the state of the volume of the sample was not monitored in the experiments of Refs. 1 and 2, in spite of the appreciable deviations of the processes in the surface layer from theoretical predictions. The experiments on tin carried out by us earlier<sup>[4]</sup> cast doubt on the validity of this seemingly obvious assertion. A transition of the interior of tin samples to the normal state was observed in Ref. 4 in a constant external field  $H < H_c$  under the action of a low-frequency alternating field, for which  $\delta_{sk} \ll d$ , where  $d$  is the thickness of the sample. We present below the results of a more detailed study of this phenomenon, performed on samples of tin and indium.

## MEASUREMENT METHOD

In the measurements, processes occurring near the surface of the sample and the state of its interior were monitored simultaneously. For observation of processes at the surface, we used the method of a transformer with a superconducting core.<sup>[2]</sup> A coil of inductance  $L_1$  was wrapped around the sample with a small gap between them, and served as the primary winding of the transformer and produced an alternating field of frequency  $\omega$  ( $30 \text{ Hz} \leq \omega/2\pi < 2 \cdot 10^5 \text{ Hz}$ ) with amplitudes up to 70 Oe. The magnitude of the alternating field was determined from the current through the coil, which was found by measurement of the voltage across a standard resis-

tance  $R$  (see Fig. 1). A second inductance  $L_2$  was put on over the first inductance coil and served as the secondary winding of the transformer. There was a parasitic sinusoidal signal in the secondary winding, due to the presence of the gap between  $L_2$  and the sample. To cancel this signal, a compensating transformer was introduced into the circuit. Satisfactory compensation was obtained at frequencies below 10 kHz. The signal from the secondary coil was observed on the screen of an oscilloscope, using either a time sweep or a sweep from the current in the circuit of the primary winding. Since the amplitude of the alternating field was determined from the current through the coil  $L_1$ , it was necessary to see to it specially that all the remaining inductance coils were open circuited to the alternating current.

The circuit monitoring the state of the interior of the sample, was also essentially a transformer, with the sample as the core, but operating at infralow frequencies. The first winding of this transformer was the superconducting solenoid, which supplied a slowly and uniformly increasing ( $\partial H/\partial t = \text{const} = 0.05 \text{ G/sec}$ ) magnetic field. The signal received by the coil  $L_3$ , passed through a filter with time constant  $\sim 5 \text{ sec}$  and after amplification was applied to the  $Y$  coordinate of an  $x$ - $y$  recording potentiometer. The superconducting transition was revealed by the emf appearing in the coil  $L_3$  when the magnetic field was pushed out at the instant of the transition from the normal state to the superconducting

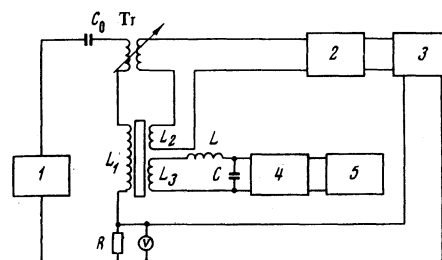


FIG. 1. Block diagram of apparatus: 1—audio frequency generator, 2—broadband amplifier, 3—oscilloscope, 4—dc amplifier, 5— $x$ - $y$  recording potentiometer.

Table I.

(Sample)	d, cm	h, cm	$\frac{d_{geo}}{d_0}$	$\beta^*$
Sn 1	0.93	3.0	$10^3$	$90^\circ$
Sn 2	0.9	5.8	$10^3$	$83^\circ$
In 1	0.9	5.8	$3 \cdot 10^3$	$19^\circ$
In 2**	0.75	5.0	$2 \cdot 10^3$	—
In 3	0.4	9.6	$10^3$	$90^\circ$

\* $\beta$  is the angle between the cylinder axis and  $C_4$ .

\*\*In 2 is a large-block polycrystal.

state or penetrated into the sample in the reverse transition. Owing to the constancy of the rate of change of the magnetic field  $\partial H/\partial t$ , the received signal, being proportional to  $\partial\Phi/\partial t$  ( $\Phi$  is the flux of magnetic induction through the sample), was proportional to  $\partial\Phi/\partial H$ . It must be noted that the decay times of the eddy currents in the best of our samples were of the order of minutes, and the transition of the sample from the superconducting state to the normal took even longer. The sweep rate was so chosen that the shape of the experimental curves did not depend on the rate.

The time of a single recording amounted to 40 min; therefore, long-term stabilization of the temperature in the cryostat was required. At temperatures above the  $\lambda$  point, of helium, stabilization was achieved by ordinary mechanical manual control; below the  $\lambda$  point, an electronic circuit was used for the stabilization. A thermocouple MT-6 built into a dc bridge was used as the pressure pickup. The unbalance voltage of the bridge was fed to a power amplifier at the output of which was a heater immersed in the liquid helium. Such a circuit maintained constant pressure in the cryostat for long times with accuracy to 0.1 mm Hg.

Single-crystal cylinders of tin and indium served as the samples. All of them, with the exception of In 2, were circular cylinders. The data on the samples are shown in Table I. The sample In 2 was elliptical in cross section; in the table, the geometric mean of the axes of the ellipse is given in place of the diameter.

The length of the coil  $L_1$ , as a rule, exceeded the height of the cylinder  $h$ , and the detecting coils  $L_2$  and  $L_3$  were one half the length of the sample. To eliminate the effect of regions of the intermediate state on the ends of the sample, several experiments were completed with the use of a short coil  $L_1$  such that its length was less than  $h$ . The results obtained with the short coil did not differ qualitatively from the previous measurements.

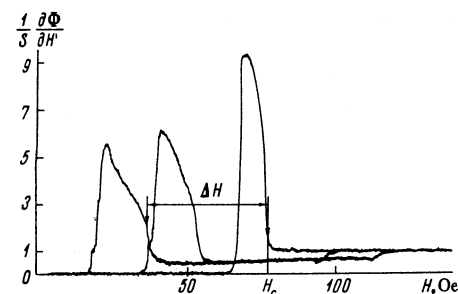


FIG. 2. Experimental recording for the sample Sn 1 at  $T = 3.22$  K.

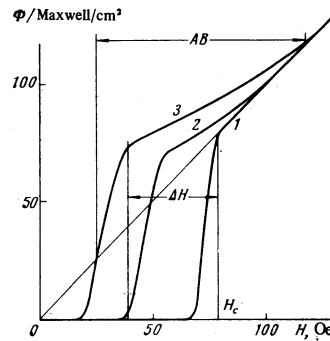


FIG. 3. Result of numerical integration of experimental curves: 1— $H_\infty = 0$ ; 2—45.5; 3—67.5 Oe.

## EXPERIMENTAL RESULTS

1. Upon introduction of the low-frequency alternating field of high amplitude, a forcing of the magnetic induction flux inside the sample was observed. This appeared in the experimental curves (Fig. 2) as a shift in the superconducting transition throughout the interior of the sample towards weaker magnetic fields. By numerical integration of the transition curves (Fig. 3), it could be established that the shift was due precisely to the increase of the magnetic flux in the sample. As is seen from Fig. 3, in the presence of an alternating field, there is a wide range of fields  $AB$  in which the magnetic induction inside the sample is significantly greater than the intensity of the external magnetic field.

2. For a quantitative description of the dependence of the injection process of the magnetic flux on the various parameters, it was convenient to use the point that is most sharply pronounced on all the experimental curves, corresponding to the transition of the entire interior of the sample to the normal state. In other words, that point at which a flux equal to  $H_c d^2/4$  is forced inside the sample. In Fig. 2 these points are marked by the arrows and a quantity  $\Delta H$  characterizing their distance from  $H_c$  is introduced.

The  $\Delta H(H_\infty)$  dependence had a clearly expressed threshold character (Fig. 4). It was linear up to those values of  $H_\infty$  at which the forward edge of the transition curve reached the point  $H = 0$ .

Changes in temperature, frequency of the alternating field and the quality of the sample all affected the value of the threshold amplitude  $H_c^L$ . The temperature dependence of  $H_c^L$  is shown in Fig. 5. As is seen from the drawing,  $H_c^L$  is proportional to  $H_c$  over a wide region of temperatures. Comparison of the results obtained on samples of different shape (Sn 1, and Sn 2; In 1 and In 3)

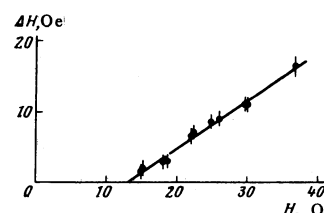


FIG. 4. Plot of  $\Delta H(H_\infty)$  for the sample Sn 1,  $f = \omega/2\pi = 1.06$  kHz,  $H_c = 92$  Oe.

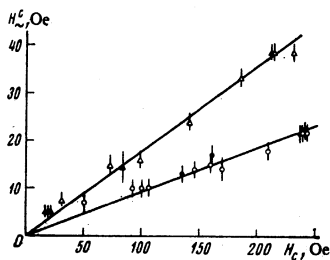


FIG. 5. Dependence of  $H_c^c$  on  $H_c$ :  $\circ$ —Sn 1;  $\bullet$ —Sn 2;  $\triangle$ —In 1;  $\blacktriangle$ —In 3;  $\omega/2\pi = 1.06$  kHz.

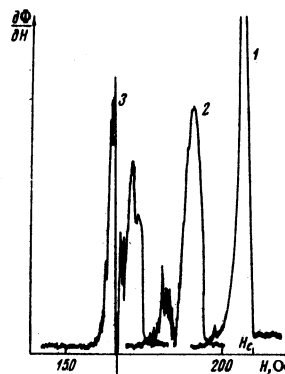


FIG. 7. Typical recording of the curve of the superconducting transition in In 3: 1— $H_{\sim} = 22.7$ ; 2—45.5; 3—67.5 Oe.

and quality (In 2 and In 1), showed that the proportionality coefficient does not depend on the shape of the sample, but is sensitive to its quality. A departure from direct proportionality between  $H_c^c$  and  $H_c$ , which somewhat exceeds the experimental error, is observed only at small  $H_c$ . Under equal conditions, the quantity  $H_c^c$  in tin was smaller, by a factor of 1.9, than in pure indium. The slope of the lines  $\Delta H(H_{\sim})$  depended weakly on the temperature. When the temperature changed from  $T = 3.58$  K ( $H_c = 20$  Oe) to  $T = 1.72$  K ( $H_c = 240$  Oe), the slope in tin decreased by a factor of 1.5.

Increase in the frequency of the alternating field led, as is shown in Fig. 6, to some increase in  $H_c^c$ . (In Ref. 4, because of the large error in the determination of  $H_c^c$ , this weak frequency dependence was not observed.) On some samples (Sn 1, Sn 2) a weak increase in  $H_c^c$  continued to a frequency of  $\omega/2\pi = 100$  kHz, above which the measurements were made difficult by the significant heating of the sample; on others (In 3) the increase in  $H_c^c$  ceased at  $\omega/2\pi \approx 10^4$  Hz. However, in that region where the increase was still observed the frequency dependences of  $H_c^c$  were close together for all samples. If it is attempted (as is done in the figure) to represent the frequency dependence in this region in the form  $H_c^c \sim \omega^n$ , then we obtain for  $n$  the value  $0.2 \pm 0.05$ .

Inclination of the magnetic field relative to the axis of the cylinder greatly weakened the injection of magnetic flux into the sample. Using, as before,  $\Delta H$  for the quantitative description, we found that upon inclination of the magnetic field a decrease occurs in the angle between the line  $\Delta H(H_{\sim})$  and the abscissa at an unchanged value of  $H_c^c$ . As an example, we give the values for the sample Sn 1. Deviation of the magnetic field by an angle of  $10^\circ$  from the axis of the cylinder decreased the quantity  $\partial(\Delta H)/\partial H_{\sim}$  by a factor of 4, by a factor of 6 and upon further inclination to  $20^\circ$ .

3. The shape of the curve of the superconducting transition changes in the alternating field, and with de-

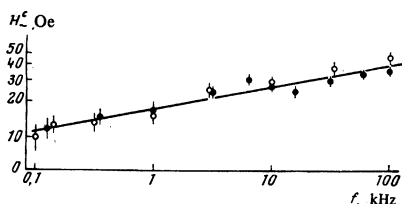


FIG. 6. Dependence of  $H_c^c$  on the frequency  $f = \omega/2\pi$  in tin samples:  $\circ$ —Sn 1;  $\bullet$ —Sn 2;  $H_c = 210$  Oe.

crease in the temperature, the deformation of the transition curve increased. In samples of tin and comparatively dirty indium (In 2), the shape of the transition curve was not very much distorted (Fig. 2), although the width of the transition was no longer determined by the geometrical dimensions of the sample, and depended on the quantity  $H_{\sim}$ . A more significant deformation of the transition curve was observed in pure indium (Fig. 7). At certain amplitudes of the alternating field, the transition to the normal state was even a two-step one. As is seen from Fig. 7, in the alternating field, in the transition region, noise with frequency of the order of fractions of a hertz increased significantly. The appearance of such noise, indicating the presence of some sort of nonstationary processes, was observed on all the samples studied (in Fig. 2, the noise is added by electronic means).

In samples for which the distortion of the transition curve was considerable (In 1, In 3), as soon as  $H_{\sim} < H_c^c$ , a broadening of the curve appeared due to a shift in the initial transition point. The accuracy of our experiments did not enable us to determine whether there was a single characteristic value of the amplitude of the alternating field for these samples, in the increase of which a shift in the initial point develops, or whether the deformation of the transition curve increases gradually.

4. The observation on the screen of the oscilloscope (Fig. 8) of transitions from the normal state to the superconducting and the reverse, which occur near the surface of the sample, demonstrated the absence of a significant dependence of the processes in the surface layer on the state of the interior of the sample. At the same time, the presence of transitions at the surface appears to be a necessary condition for the injection of

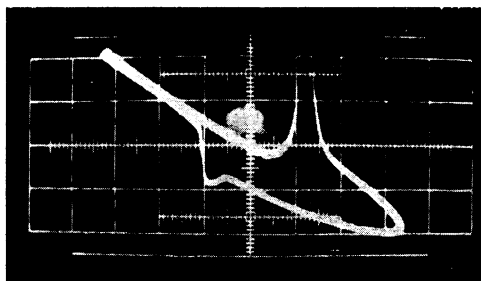


FIG. 8. Example of photograph of the screen of the oscilloscope; Sn 1,  $\omega/2\pi \approx 300$  Hz,  $H_c = 72$  Oe,  $H_{\sim} = 45$  Oe,  $H = 59$  Oe.

the flux. It was noted that the origin of the transition curve to the normal state of the entire interior of the sample always coincided with the appearance of transitions in the surface layer, and their disappearance took place at the point where the curve  $\Phi(H)$  reached the line  $\Phi = H\pi d^2/4$ .

The increase in the amplitude of the alternating field led to the increase of hysteresis in the transitions at the surface, principally because of the increase in the magnetic supercooling.

Establishing the external magnetic field  $H = H_c$  and, by means of the alternating field, transforming the interior of the sample to the normal state, we could estimate the thickness of the superconducting layer  $\delta_1$  which forms at the surface from the picture on the screen of the oscilloscope, and obtain the ratio

$$\frac{\delta_1}{\delta_{sk}} = \frac{S_1 \pi H_{\sim}}{2(S_n - S_s) H_c},$$

where  $S_1$  is the area under the transition curve and  $S_n$  and  $S_s$  are the areas of the ellipses on the screen of the oscilloscope in the normal and superconducting states, respectively. For Sn 1, we thus obtain  $\delta_1/\delta_{sk} \approx 0.12$  from Fig. 8. The same figure is obtained for In 1 at  $H_c = 125$  Oe,  $H_{\sim} = 60$  Oe,  $\omega/2\pi = 500$  Hz.

## DISCUSSION

We present below arguments that enable us to understand at the qualitative level why the magnetic flux inside a sample can be greater than  $H\pi d^2/4$  in an alternating field, and to estimate the value of the flux pushed into the sample. The basic difference of the experimental conditions from those used in the calculation of Lifshitz<sup>[3]</sup> lies in the periodic appearance of a superconducting layer near the surface of the sample. This layer cuts off the layer of normal metal from the surface. Just this difference is used below for explanation of the experimental results.

It is convenient to begin the discussion with the case of very low frequencies  $\omega \ll c^2/\sigma d^2$ , which has no direct relation with the experiment, but which satisfies the condition  $\omega^{-1} \ll \tau_M$ . Here  $\tau_M$  is the time in which the magnetic flux is ejected from the sample in the transition to the superconducting state,  $\tau$  is the conductivity of the metal. As is well known,<sup>[5]</sup> the time  $\tau_M$  is rather large. According to our estimate,  $\tau_M \sim 3 \times 10^2$  sec for a sample of thickness 1 cm. The reverse transition to the normal state takes place rapidly within the time<sup>[6]</sup>

$$\tau \sim \frac{\sigma d^2}{c^2} \frac{1 + \alpha}{\alpha},$$

where  $\alpha$  is the supercriticality of the magnetic field ( $\tau \sim 50$  sec at  $\sigma = 10^{22}$  cgs esu,  $\alpha \sim 1$ ). At frequencies with which we are concerned,  $\delta_{sk} \gg d$ , and the field in the interior of the sample is homogeneous and is identical with the external field at  $H + H_{\sim}(t) > H_c$ . If  $H + H_{\sim}(t) < H_c$ , then the field inside is equal to  $H_c$ . In average over a long interval of time, the field in the interior is equal to

$$H_0 = H_c + \frac{(H - H_c)\varphi}{\pi} + \frac{H_{\sim}}{\pi} \sin \varphi,$$

$$\varphi = \arccos \frac{H_c - H}{H_{\sim}}.$$

The transition of the sample to the normal state takes place in an external magnetic field equal, say, to  $H_c - H_{\sim}$ . The mean field in the sample  $H_0 > H_c$  even at  $H < H_c$  and this difference is connected with the valve-like action of the superconducting crust that forms on the surface of the superconductor at  $H + H_{\sim}(t) < H_c$ . (Here we have not taken into account the magnetic supercooling. Its role is discussed below.)

For our samples, however, the opposite relations  $\omega^2 \gg c^2/\sigma d^2$ ,  $\delta_{sk} \ll d$  were satisfied. As an example, we give these relations for tin. The skin depth in it under conditions of the anomalous skin effect was measured in Ref. 7. Conversion of the data there to the frequency of  $10^3$  Hz, at which most of our experiments were carried out, gives a value  $\delta_{sk} \approx 4 \times 10^{-3}$  cm. Calculation according to the anomalous skin effect is valid if  $\delta_{sk} \ll l$ , where  $l$  is the free path length of the carrier. The quantity  $l$ , obtained from the resistance ratio  $\rho_{300}/\rho_0$ , amounted to  $3 \times 10^{-2}$  cm in our experiments, so that  $\delta_{sk} \ll l \ll d$ .

If, as before, we do not take magnetic supercooling into account, then we can obtain in this case the formula given for  $H_0$  above. Continuity of the tangential component of the magnetic field requires that the field in the surface layer of a normal metal track the external. Therefore, upon decrease in the external field below  $H_c$ , we can expect the appearance of a thin superconducting sheath in the surface layer. This sheath, being superconducting, should be penetrable for the magnetic flux, at least until the field in the volume of the sample  $H_0 > H_c$ . Similar sheaths, capable of assuring jumps in the tangential component of a magnetic field of intensity  $H_c$  at distances of the order of the coherence length  $\xi$  are known as the two-dimensional mixed state of type I superconductors and were first observed experimentally by I. Landau and Sharvin.<sup>[8,9]</sup> Their theoretical description is given in Refs. 10 and 11. It is essential for us that after the onset of the sheath of the two-dimensional mixed state, the magnetic field at distances of the order of  $\xi$  from the surface of the sample remains equal to  $H_c$ , while the external field is less and the internal field greater than  $H_c$ . When the external field, increasing, again becomes equal to  $H_c$ , the layer of the two-dimensional mixed state disappears and the field on the surface will be equal to the external field. In the mean, over a long interval of time, the field at a distance of the order of  $\xi$  from the surface corresponds to the formula for  $H_0$  given above. The same will naturally be the average field in the interior of the sample. Figure 9 shows a comparison of the  $H_0(H)$  dependence with the experimental curve.

Magnetic supercooling can significantly change the course of the processes in the surface layer. An observation of the picture on the screen of the oscilloscope has shown, the nuclei of the superconducting phase which form on the surface of the sample, because of the presence of supercooling, grow into the interior to a

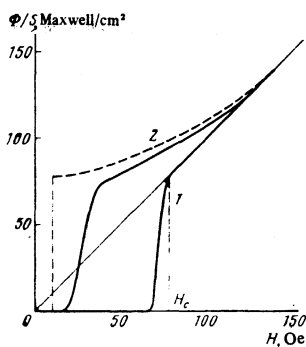


FIG. 9. Comparison of the experimental curves (Sn 1—continuous line) with the calculated (dashed): 1— $H_0 = 0$ ; 2—67.5 Oe.

depth  $\delta_1 \sim 10^{-3}$  cm and, after merging, create a superconducting layer of thickness  $\delta_1 \gg \xi$  around the middle part of the sample. A magnetic flux  $\Phi \sim \pi d \delta H_c$  is then forced out of the sample. (The growth of the nuclei of the superconducting phase and the forcing-out of the magnetic flux were investigated in detail in Ref. 12.) After formation of the superconducting layer, the change in the magnetic flux in the samples decreased so much that it became unobservable at our accuracy after a time  $\sim 1/\omega$  (Fig. 8).

If we assume that  $H_0 > H_c$ , then the inner boundary of the superconducting layer should begin to move towards the surface of the sample and reach it within a time

$$\tau_1 \sim \left( \frac{\delta_1}{\delta_{sk}} \right)^2 \frac{H_c}{\omega(H_0 - H_c)}.$$

At small supercriticalities we have  $\omega\tau_1 > 1$  and the boundary of the superconducting layer generally does not reach the surface within one period of the alternating field. In this case, the magnetic flux inside the sample is preserved and the magnetic field at distances from the surface exceeding  $\delta_1$  will be as before either be greater than or equal to  $H_c$ . This means that even in the presence of a macroscopic superconducting layer at the surface forcing of the magnetic flux into the sample can be observed.

At large supercriticalities we have  $\omega\tau_1 < 1$ , so that the boundary of the superconducting layer manages to reach the surface within one period and form a layer there of a two-dimensional mixed state. Precisely this combined case evidently corresponds to the experimental situation. A weak frequency dependence can be connected with the relation of the time of existence at the surface of the macroscopic superconducting layer and the sheath of the two-dimensional mixed state.

The value of the magnetic supercooling determines the critical value of the amplitude of the alternating field  $H_c^c = (H_c - H_{cm})/2$  (here we understand by  $H_{cm}$  the experimental value of the field at which the formation of nuclei of the superconducting phase begins on the surface). Actually, at  $H_c < (H_c - H_{cm})/2$ , the calculation of Lifshitz<sup>[3]</sup> is valid and the interior of the sample remains superconducting, while  $H < H_c$ . As was noted in Ref. 4, the transitions at the surface can be observed even at  $H_c < H_c^c$ , probably because of the presence of defect spots in which  $H_{cm}$  is greater than the average on the surface. A closed superconducting layer arises at the surface only at  $H_c = H_c^c$ . The linear dependence of  $H_{cm}$  on  $H_c$  is explained by the proportionality of  $H_c^c$  and  $H_c$ .

It would be interesting to carry out similar measurements on superconductors for which the Ginzburg-Landau parameter is  $0.42 < \kappa < 2^{-1/2}$ , for example on lead. In these superconductors, the  $\Delta H(H_c)$  dependence should have a threshold character. Therefore it can be hoped that the experimental and theoretical curves will be in better agreement.

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