## QUANTIZATION OF MAGNETIC FLUXES IN SIMPLY CONNECTED SUPERCONDUCTING CYLINDERS

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Results are presented of an experimental investigation of quantization of the magnetic flux in simply connected superconducting cylinders. The samples (filaments with diameters of the order of a micron) are prepared from InPb containing from 0 to 5 at.% of Pb, which corresponds to a transition from type I to type II superconductors. The phase diagram of the cylinders in a parallel field has an oscillating shape. The oscillation period decreases with reduction of the temperature. The oscillation amplitude increases with decrease of the sample cross section. The quantum oscillations remain in samples with H < 0.4 and in pure In samples. This is ascribed to supercooling of the samples below the thermodynamic critical field. Qualitative agreement is observed with existing theoretical works on quantum transitions in continuous cylinders with surface superconductivity.

WE have previously reported in <sup>[1]</sup> the observation of oscillations of the resistance of singly-connected superconducting cylinders of small diameter upon variation of the magnetic field parallel to the cylinder axis near  $T_c$ . The observed phenomenon was attributed by us to quantization of the magnetic flux by the surfacesuperconductivity currents. A theoretical calculation of the electric resistance of a solid cylinder in the state of surface superconductivity, carried out by M. G. Khamudillin, <sup>[2]</sup> gives an oscillating transition curve that coincides with the R(H) curves given in our paper.<sup>[1]</sup> We present here the results of further investigations of quantum transitions in singly-connected cylinders.

## SAMPLES AND EXPERIMENT

The samples were thin wires prepared by drawing a glass capillary with the molten metal. The glass was not removed from the sample. The metal under the glass has a mirror surface with a small number of shallow pits, constituting several percent of the total sample. surface. The sample diameter d, ranging from 1 to several microns, was estimated with the aid of an optical microscope and was determined more accurately by measuring the resistance at room temperature. The mean free path at 300 °K is of the order of  $10^{-6}$  cm. which is smaller by two orders of magnitude than the sample diameter, and consequently the error in the determination of the diameter from the resistance at room temperature is negligible. The wire was soldered to the sample with pure In. A dc current of several microamperes was made to flow through the sample, corresponding to a current density on the order of  $10 \text{ A/cm}^2$ . A voltage proportional to the wire resistance was applied to one coordinate of an x-y recorder, and a voltage proportional to the magnetic field was applied to the other coordinate.

In a number of cases, dR/dH was recorded automatically. In this case the modulating magnetic field was produced by an additional coil. The modulation amplitude was 0.1 Oe. The temperature was stabilized by a manostat with accuracy not worse than  $10^{-4}$  °K (in the region 3-4 °K).

The investigated material was an InPb alloy. The Pb content in the alloy was chosen such that the parameter  $\kappa$  varied in a wide range. Alloys containing more than 4 at.% are type II superconductors and those with less than 2 at.% are type I superconductors with  $\kappa \leq 0.4$ , i.e., the case  $H_{c3} < H_c$  is realized. Alloys with Pb concentration from 2 to 4 at.% have accordingly  $0.4 \leq \kappa$  $\leq$  0.7. Table I shows the characteristics of the investigated samples. The mean free path at  $4.2 \,^{\circ}\text{K}$  ( $l(4.2 \,^{\circ}\text{K})$ ) was calculated from the relation  $\rho l = 1.4 \times 10^{-11}$  ohm $cm^{2}$ <sup>[4]</sup> under the assumption that  $\rho l$  remains the same also for the InPb alloy at small Pb concentrations. The value  $\xi_0 = 2400$  Å for pure In was taken from <sup>[5]</sup>. The values of  $\xi(0)$  for a series of samples with Pb impurities were calculated with allowance for the influence of the mean free path on the coherence length.<sup>[6]</sup>

## RESULTS AND DISCUSSION

The series of R(H) curves plotted at different temperatures for a sample of 3.8  $\mu$  diameter containing

Table I. Characteristics of investigated samp	les	*
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Pb content, at.%	ρ(300° K), 10 <sup>-6</sup> ohm-cm	$\frac{\rho(300^{\circ} \text{ K})}{\rho(4,2^{\circ} \text{ K})}$	<i>l</i> (4,2° K), 10 <sup>-6</sup> cm	ξ., Å	<sup>d</sup> , 10 <sup>-4</sup> cm
0	8.64	220	356	2400 [5]	2.8
1	9,81	15.4	22	<b>163</b> 0	1.8
2	11.65	$^{8.2}_{9}$	9.85 10.8	1370 1410	4.2
2,5	12.8	$\begin{array}{c} 6.85 \\ 6.6 \\ 7.05 \\ 6.9 \\ 7.4 \\ 7.3 \end{array}$	7.5 7.2 7.7 7.6 8.1 8.1 8	1200 1170 1220 1200 1280 1280 1280 1260	2 2.3 3.2 3.5 3.9 5.1
3	13	5.88	6.25 5.3	1070 1020	4
5	17.5	4.76	3,8	890	5,0

\* $\rho(300^{\circ}K)$  and  $\rho(4.2^{\circ}K)$  are the resistivities at 300 and 4.2°K, respectively.



FIG. 1. Dependence of the resistance on the parallel magnetic field of a cylinder of In + 2.5 at.% Pb with diameter  $3.8\mu$ .



FIG. 2. Series of R(H) curves plotted at equal temperature intervals. The initial and final temperatures are 3.611 and 3.145°K, respectively. The arrows indicate the locations of the oscillations.

2.5 at.% Pb is shown in Fig. 1. With decreasing temperature, resistance oscillations appear on the transition curve. The period of the oscillations, in terms of the magnetic field, decreases in inverse proportion to the cross section of the sample, and several bursts (oscillations) of the resistance are observed for the largediameter samples (Fig. 1). With decreasing sample diameter, the period of the oscillations increases and may equal or even exceed the width of the smearing of R(H). Figure 2 shows the transitions of the sample  $In_{97.5}$ -Pb<sub>2.5</sub> of 2  $\mu$  diameter in a magnetic field, measured at equal temperature intervals. The positions of the oscillations are marked by arrows. The density of the curves also oscillates with a period equal to a distance between the arrows.

The series of transition curves plotted in an isothermal regime with small temperature intervals makes it possible to construct the phase diagram  $H_c(T)$ . The curves were plotted for different criteria for the selection of the critical field.  $H_c$  was chosen to be the field at which  $R/R_N = 0.99$ , or else 0.5, or finally 0.01.

The  $H_c(T)$  plot shown in Fig. 3 is for a sample of 2  $\mu$  diameter. Oscillations are clearly seen, and their number reaches 10–15 for certain samples. The oscillating part of the curve is separated in the form of oscillations of the critical temperature, with amplitudes that reach 10<sup>-2</sup> °K for a sample having this diameter.

The singularities of the  $H_c(T)$  curves obtained for different criteria occur at the same magnetic field, indicating that the effect is insensitive to the choice of the critical-field criterion. In magnetic fields perpendicular to the cylinder axis, the phase diagram turns out to be smooth and has no oscillations.

The following characteristic features of the effect should be noted:

Oscillations of the critical temperatures were retained in samples of alloys with  $\kappa < 0.4$  and in samples of pure In.

2. When  $\kappa < 0.4$ , hysteresis of the curves in increasing and decreasing fields becomes noticeable. For the



FIG. 3. Phase diagram  $H_c(T)$  of a sample of In + 2.5 at.% Pb of  $2\mu$  diameter. The oscillating parts of the phase diagrams are shown separately for critical fields corresponding to 0.5 R<sub>N</sub> and R<sub>N</sub>. The H<sub>c</sub>(T) curve is smooth in a perpendicular magnetic field.

Table II.								
$n_1$ (initial)	0	1	2	34				
$n_2$ (final)	1	2	3					
$\frac{d^2}{\xi^2}(t)$	7	13.4	19.5	24.8				
$\xi$ (0), Å	1080	1070	1080	1090				

In samples, oscillations were observed only on the curve obtained in decreasing fields. For samples with  $\kappa \sim 0.4$ , the oscillations are more strongly pronounced in decreasing fields. The amplitude  $\Delta T_C$  of the oscillations depends on the orientation of the sample in the magnetic field. In oblique fields, it decreases sharply, and practically vanishes at angles near 15° (Fig. 4).

In the theoretical papers, Saint James,  ${}^{(7)}$  Dolmasso and Pagiola,  ${}^{(B)}$  and also Khamidullin  ${}^{(9)}$  determined those values of the ratio  $d^2/\xi^2(t)$  (t is the relative temperature), at which one observes in singly-connected cylinders transitions from one state to another, differing by a quantum of the flux  $\Phi_0$ . For example, at  $d^2/\xi^2(t) = 7$  there is a transition from a state with n = 0to a state with n = 1 (one flux quantum in the cross section); at  $d^2/\xi^2(t) = 13.4$  there is a transition from n = 1to n = 2, etc. On the phase diagram, quantum transitions from the state n to the state n + 1 corresponds to points with maximum deviations from the envelope of the plot of the critical field  $H_c(T)$ . Knowing the sample diameter and the temperatures at which the transitions are observed, it is possible to determine the values of  $\xi_0$ .

Table II shows the values of  $\xi_0$  obtained for a number of succeeding transitions  $n \rightarrow n + 1$ , for the sample whose phase diagram is shown in Fig. 3.

The values of  $\xi(0)$  obtained in Table II agree with the values of  $\xi(0)$  given in Table I for samples having the same Pb content. Therefore the value  $\xi_0 = 2400$  Å for pure In also agrees with our experimental data. A similar value of  $\xi_0$  was obtained for pure In by McLach-lan<sup>[10]</sup> in an investigation of the susceptibility of solid cylinders of an InBi (1.84 at.%) alloy. The values  $\xi_0 = 4400$  Å encountered in the literature for In,<sup>[4, 11]</sup> are apparently significantly overestimated.

A relation describing the temperature dependence of the period of the oscillations was obtained by Kulik and



FIG. 4. Dependence of the oscillation amplitude ( $\Delta T_c$ ) on the sample position in the magnetic field.  $\varphi$ -angle between the sample axis and the magnetic-field direction.

FIG. 5. Temperature dependence of the period of the oscillations of a sample of In + 2.5 at.% Pb of  $2.3\mu$  diameter. The cross corresponds to the values n = 1 and  $\gamma$  = 0, and the points to n = 0, 1, 2, ..., and  $\gamma$  = 0.67. The solid curve is theoretical and obtained from (1) at  $\xi(0) = 1.2 \times 10^{-5}$  cm for the given sample, t = T/T<sub>c</sub>.

has the following form: [12]

$$\pi [r - 0.6\xi(t)]^{2} H = (n + \gamma) \Phi_{0}, \qquad (1)$$

where  $\gamma = 0.67$ . It is seen from (1) that the period of the oscillations in terms of the field decreases with decreasing temperature, because it is not the total crosssectional area of the cylinder which is being quantized, but only the area of the effective cross section, with a radius smaller than the cylinder radius by an amount  $0.6 \xi(t)$ .

Figure 5 shows the temperature dependence of the period of the oscillations of a sample of 2.3  $\mu$  diameter. The cross corresponds to the values n = 1 and  $\gamma = 0$ , and the points to n = 0, 1, 2, ... and  $\gamma = 0.67$ . The solid line is the theoretical plot obtained from (1).

With increasing number of flux quanta in the cylinder cross section, or with decreasing temperature, the amplitude of the critical-temperature oscillations decreases, in qualitative agreement with the conclusions of <sup>[7,8]</sup>. A theoretical calculation of the amplitude of the effect ( $\Delta T_c$ ), carried out by Kulik for large quantum numbers n, leads to a formula<sup>[12]</sup> analogous to the Parks-Little effect:<sup>[13]</sup>

$$\Delta T_{c} = 0.14 T_{c} (\xi_{0}/r)^{2}.$$
 (2)

Figure 6 shows the dependence of  $\Delta T_c$  on  $r^{-2}$  for a series of samples with the same Pb content. In agreement with relation (2), the amplitude of the effect increases linearly with decreasing sample cross section. For the first oscillations (small n), the experimental points lie above the theoretical plot. With increasing n (or with decreasing temperature), the amplitude  $\Delta T_c$  decreases, approaching relation (2), and for the fifth and all succeeding oscillations are lower than those predicted by (2).

Thus, the experimental data obtained by us agree with the theoretical papers<sup>[7-9]</sup> dealing with quantization in singly-connected cylinders in the surface-superconductivity state. The existence of oscillations of  $T_c$ in alloys with  $\kappa < 0.4$  and in pure In is apparently due to supercooling of the samples below the thermodynamic



FIG. 6. Plot of  $\Delta T_c$  against  $r^{-2}$  for a series of samples of InPb + 2.5 at.% Pb.  $\bullet$ -amplitude of first oscillation,  $\blacktriangle$ -of second oscillations,  $\bigcirc$  (near the solid line)-of fourth oscillation, +-of fifth oscillation. The heavy line shows the theoretical dependence (2) at  $\xi(0)$ , taken from Table I for each sample.

critical field.<sup>[14]</sup> This is confirmed both by the appearance of hysteresis at  $\kappa < 0.4$  and by the absence of oscillations in increasing fields (a detailed article will be published). In the latter case the state with n = 0 is conserved in the sample up to the critical field value. Since the supercooled field in samples with  $\kappa < 0.4$  has an oscillatory temperature dependence, it can be assumed that the superconductivity in the supercooling field originates on the sample surface, forming a doubly-connected region. This agrees with the conclusions of Faber, <sup>[15]</sup> who investigated the processes of generation of a new phase in type I superconductors. He believed that the nuclei of the superconducting phase, arising near the surface, form annular shells before they begin to penetrate into the interior and fill the entire sample.

It can be concluded from the foregoing experimental data that the observed effect agrees with the model for the quantization of a magnetic flux by surface-superconductivity currents, and that the physical cause of the oscillations is the existence of circulating currents on the cylinder surface, connected with the general condition for the fluxoid quantization and varying periodically with the magnetic field.

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