QUANTUM OSCILLATIONS IN THE SUSCEPTIBILITY OF A SUPERCONDUCTING TIN MICROCYLINDER

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The susceptibility of a thin-walled microcylinder is measured as a function of magnetic field at various temperatures, and oscillations due to fluxoid quantization are observed. In order to explain the magnetization close to T_C, where the phase transition is of second order, the expression for the flux in a thin-walled microcylinder derived by several authors must be used to relate the observed increments in magnetization to the absolute quantum of the fluxoid.

The quantization of flux was first predicted by London[1] and subsequently Deaver and Fairbank[2] and independently Doll and Näbauer[3] verified that the flux trapped in superconducting microcylinders was indeed quantized in units of $hc/2e$. This was followed by a large number of theoretical papers on the subject (see Douglass[4] for a complete bibliography). It has also been observed[5-7] that the transition temperature of a thin-walled microcylinder is periodic, with period $hc/2e$, in the applied magnetic flux threading the cylinder. The effect was first explained theoretically by Tinkham.[8] Flux quantization has most recently been observed by Lischke[9] using an electron interferometer.

In the present experiment the susceptibility of a microcylinder is measured. The cylinder was prepared by evaporating 2200 Å of tin onto a rotating glass fiber (radius a). The thickness of the tin (d) was estimated by dividing the thickness of tin deposited on an adjacent glass slide (measured by optical interference) by π. After completion of the experiment a Cambridge scanning electron microscope was used to measure the thickness of the tin-coated fiber and a result of $2a+2d=5.6\pm0.2$ μ was obtained. The 2-mm-long microcylinder, protected by a coat of lacquer, was placed in a mutual inductance, the primary of which gave a field uniform to 10% over the microcylinder, and the secondary of which consisted of two series-opposed coils of 20 turns each and an effective diameter of 180 μ. The detection system,[10] operating at 140 000 Hz, was the one previously used to study superheating and cooling in single spheres[11,12] and whiskers,[13] with the addition of a simple canceling network to buck out the background signal from the mutual inductance to about 1 part in 10^4. The off-balance signal of the detection system was recorded as a function of magnetic field of an X-Y recorder. The temperature was stabilized[14] during the recordings to better than 0.1 mK and measured using a calibrated germanium thermometer, which allowed relative temperature measurements of 0.2 mK. The earth's field was canceled to better than 0.02 G.

Recordings of the off-balance signal versus magnetic field for a 0.1-G peak-to-peak alternating magnetic field are shown in Fig. 1 at various temperatures. Similar recordings are obtained for peak-to-peak fields from as small as the signal-to-noise ratio would permit to 0.3 G. At 0.3 G the recordings are somewhat smeared out. Oscillations with a period of $H_\varphi=0.905\pm0.005$ G appear at all temperatures and close to T_C the differential susceptibility is actually paramagnetic. The recordings in Figs. 1(a) and 1(b) are reversible, while Figs. 1(c) and 1(d) show hysteresis. The natural conclusion that the order of the phase transition is changing is further strengthened when we note that in the plot of the difference between the zero-field and the normal-state signal versus reduced temperature, shown in Fig. 2, there is a break in the curve at $t=0.991$ indicating a possible change in the susceptibility from the normal diamagnetic shielding by supercurrents. Figure 2 also shows that the amplitude of the first paramagnetic spike has its maximum value at a slightly higher temperature. The transition temperature 3.839 ± 0.001 K was determined by extrapolating these signals to zero.

The detailed theory for the onset of the second-order phase is known for two experimental situations. They are (A) where the magnetic field is equal on both sides of the film, when the condition[15] for a second-order phase transition is that

$$d \leq 5^{1/2}\lambda(T); \tag{1a}$$

(B) where the field is zero inside and has a finite value outside or alternatively has a finite value inside and is zero outside. In the limit $d \ll a$ the condition[4,16] for a second-order phase transition is that

$$\lambda^2(T) \geq ad. \tag{1b}$$

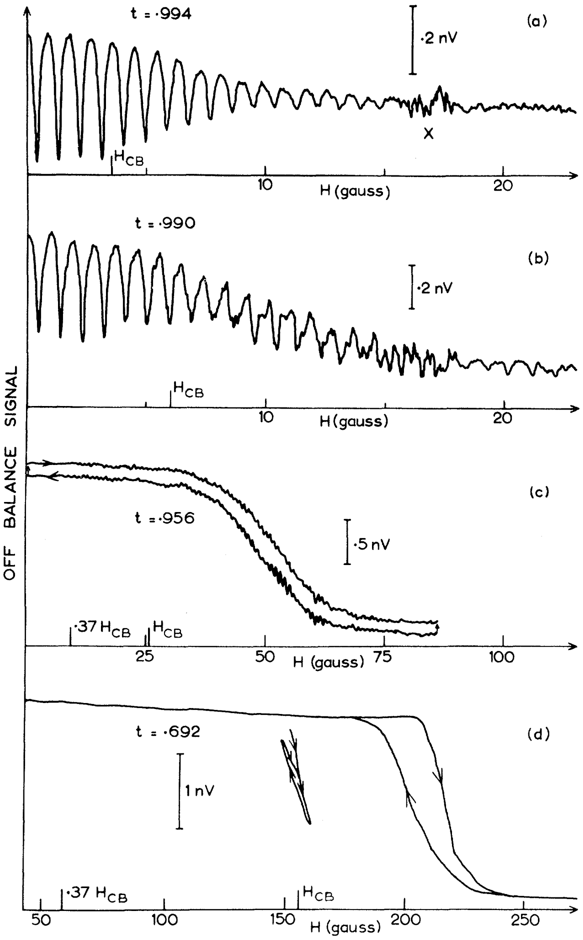

FIG. 1. Recorder tracings of the off-balance signal versus the magnetic field (H) at various values of the reduced temperature $t = T/T_C$. The bars give an indication of the vertical scale. The value of the bulk critical field is shown on the magnetic field axis. A displaced minor hysteresis loop is also shown in (d). In (c) the traces have been displaced from each other to avoid overlap of the oscillations.

Assuming $\lambda(0) = 800$ Å and $\lambda(T) = \lambda(0)(1-t^4)^{-1/2}$, a second-order phase transition in situation A requires $t \geq 0.766$, while in situation B the requirement is that $t \geq 0.997$. The present situation obviously falls between the two cases. It will later be shown that, at least where there is a second-order phase transition, the field configuration is intermediate between situations A and B.

It is obvious from the reversibility shown in Fig. 1(a) that flux is not trapped and that n, the number of fluxoids, is continually changing in order to bring the system into the lowest-energy state. Lüders[17] predicts that the number of fluxoids will be determined by the condition

$$(n-\tfrac{1}{2})\varphi_0 < \pi\bar{r}^2 H_{\text{ext}} < (n+\tfrac{1}{2})\varphi_0. \tag{2}$$

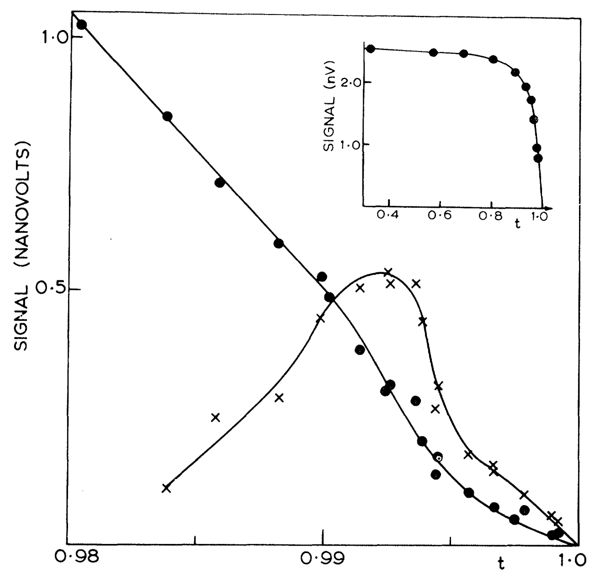

FIG. 2. The difference in the off-balance signal at zero field and when the sample is normal (closed circles), plotted against the reduced temperature $t = T/T_C$. The main figure shows the results close to T_C while the insert shows the results at lower temperatures. Also shown in the main figure is the size of the first quantum spike (×) as a function of t.

Here $\varphi_0 = ch/2e \approx 2\times10^{-7}$ G/cm^2 and $\bar{r}^2$ is given by

$$\bar{r}^2 = [(a+d)^2 - a^2]/2\ln[(a+d)/a]. \tag{3}$$

For a thin-walled cylinder ($a \gg d$) $\bar{r} \approx r_m = (2a+d)/2$. Using the measured value of $H_\varphi = \varphi_0/\pi\bar{r}^2$ a value of 2.70 μ was obtained for $\bar{r}$. This excellent agreement with the measured value (2.69 μ) should, at least at this stage, be regarded as somewhat fortuitous. Douglass[4] has suggested that the value of n should remain constant in a changing magnetic field unless the pairs can make mechanical contact with the lattice. The present experiments do not necessarily violate this condition as the finite resistivity due to the alternating currents may provide the necessary contact.

As the magnetization is easier to treat theoretically than the susceptibility, the susceptibility, in Fig. 1(a), has been integrated in Fig. 3. As the true zero of the susceptibility is not known, a constant times the magnetic field is subtracted from the integrand at every field, the constant being chosen such as to make the magnetization zero at the critical field. The critical field in Fig. 1(a) is assumed to be 17.1 G. The "noise" at the point marked × in Fig. 1(a), which is repro-

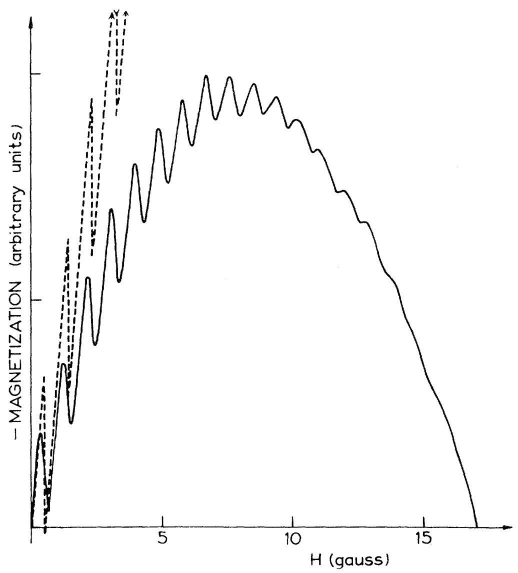

FIG. 3. The magnetization obtained by integrating Fig. 1(a) is plotted as a function of magnetic field (solid line). Also shown is the theoretical curve for M (broken line) discussed in the text.

ducible, may well correspond to different parts of the film going normal, as no more oscillations are observed above this point. As the absolute value of the susceptibility is not known either, the magnetization has to be expressed in arbitrary units. It will immediately be seen that the magnetization is always diamagnetic and never paramagnetic. The magnetization increases, with oscillations corresponding to the changing of the quantum number n, until a critical current is reached, after which the field penetrating the film lowers the value of the critical current (and magnetization) to zero. The diamagnetism is due to the fact that it is the fluxoid, given by

$$\varphi + 2\pi rc\Lambda J_\varphi = n(hc/2e), \tag{4}$$

and not the flux which is quantized. In Eq. (4) $\Lambda = m/(2e^2|\psi|^2) = 4\pi\lambda^2/c$ where $|\psi|^2$ is the number density of superconducting pairs and J is the current density which can be taken as constant for $d<\lambda$. Under these conditions φ can be expressed as[4,18-22]

$$\varphi = nhc/2e(1+2\lambda^2/rd)^{-1}. \tag{5}$$

The value of r to be used in this equation is not clearly defined by the authors.[4,18-22] A simple calculation of the flux inside a cylinder, the walls of which carry a uniform current, shows that the correct mean r to use in this case is

$$r_0^2 = [(a+d)^2 + a(a+d) + a^2]/3$$

but the difference between the value of r_0, $\bar{r}$, and r_m is again negligible and henceforth a single mean denoted by r will be used.

On substituting the value φ from Eq. (5) into Eq. (4) and Eq. (4) into Lüders's[17] expression

$$2\pi rc\Lambda J_s = n\varphi_0 - \pi r^2 H_{\text{ext}}, \tag{6}$$

the following expression is obtained for the total current J:

$$J = J_s - J_\varphi$$
$$= [n\varphi_0(1+2\lambda^2/rd)^{-1} - \pi r^2 H_{\text{ext}}]/2\pi r\Lambda, \tag{7}$$

where the value of n is determined by Eq. (2). J can be thought of as consisting of two components, J_s which has a sawtooth shape and is determined by the difference between the external flux and $n\varphi_0$, and J_φ which increases by an equal increment every time n increases. The total current density J is directly proportional to the magnetization in a thin-walled cylinder. A negative J corresponds to diamagnetism. Hence the magnetization M may be expressed as

$$M = C|\psi|^2[n\varphi_0(1+2\lambda^2/rd)^{-1} - \pi\bar{r}^2 H_{\text{ext}}]. \tag{8}$$

With $\lambda_0 = 800$ Å and $t = 0.994$, $(1+2\lambda^2/rd)^{-1} = 0.53$. The magnetization from Eq. (8) with $C|\psi|^2$ chosen so that the initial slope from Eq. (8) and the experimental value agree is shown by the dashed line in Fig. 3. The qualitative agreement between the two is excellent especially when it is realized that the value of $|\psi|^2$ and hence J decrease as the field penetrates. No attempt has been made to fit the results with a field-varying value of $|\psi|^2$ as this particular situation has not been treated theoretically. As the field inside is about one-half of the external field, it is now quite clear why the experimental results lie between situations A and B.

The critical field or the point where the oscillations cease corresponds to approximately $H_{CB}\lambda/d$. This is lower than expected for situation A, which when $\lambda > d$ gives[15]

$$H_C = 24^{1/2} H_{CB}\lambda/d, \tag{9a}$$

whereas situation B gives[16]

$$H_C = 8^{1/2} H_{CB}\lambda/a \tag{9b}$$

which is too low. H_{CB} is calculated using $H_{CB}(T) = H_{CB}(0)(1-t^2)$ together with the Mapother correction for tin using a value of 314.2 G for $H_{CB}(0)$. $H_{CB}(0)$ is calculated using the similarity principle, i.e., $H_{CB}'(0) = H_{CB}(0)T_C'/T_C$ using the values 305.5 G and $T = 3.722$°K as the correct ones for bulk unstrained tin.

Even when the sample is clearly undergoing a first-order phase transition such as in Fig. 1(d) there appear from time to time oscillations with the correct period. As both the period and the amplitude of these oscillations are too small to be seen on the scale of Fig. 1(d) no attempt has been made to show them in the figure. The oscillations in Fig. 1(d) are very similar to those shown in Fig. 1(c), where hysteresis is just becoming apparent. These oscillations usually only appear above $0.37H_{CB}$ where the recorder tracing also begins to dip slightly. This is in good agreement with the theoretical value[16] for the field at which the flux should start to penetrate in order for the sample not to be driven normal, which is $H_{\text{pen}} = H_{CB}(2d/a)^{1/2}$ for $a \gg d$. Evaluating this with $d = 2200$ Å and $a = 2.59$ μ we get $H_{\text{pen}} = 0.41H_{CB}$. As may be expected, the area of the hysteresis loop increases with decreasing temperature, i.e., as we move away from the second-order phase transition. The upper critical field is not clearly defined though it is obviously much higher than H_{CB}. The measured value is lower than the critical field expected for situation A when $t \approx 0.956$ to $t \approx 0.8$. The agreement below $t \approx 0.8$, where the definite shoulder on the hysteresis curve shown in Fig. 1(d) develops, is moderately good. (A complete plot of H_C/H_{CB} vs d/λ for situation A is given in Fig. 1 of Ref. 12.) The fact that at about $t = 0.8$, $d = 5^{1/2}\lambda$ is also probably of significance.

A minor hysteresis loop traced using the dc field is shown in Fig. 1(d). On a large enough scale quantum oscillations can clearly be seen on the whole minor hysteresis loop. The ac field also takes the sample around some hysteresis loop which is less than H_φ wide. The nature of this loop clearly depends on whether it is centered on a value of nH_φ or not, which is why oscillations in the off-balance signal appear, with the quantum period, even in the case of first-order phase transitions.

Further experiments are being carried out to study the phenomena described in this paper in greater detail.

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