## Anomalous behavior of superconducting samples with a fixed number of vortices

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We report an anomalous reversible temperature dependence of the magnetization of amorphous type-II superconductors. The experimental results were obtained studying the vortex pinning in samples with very low vortex concentration. The anomaly is shown to be a consequence of the lack of flux quantization in finite samples.

Pinning forces in homogeneous amorphous superconductors have received recent attention,  $^{1-3}$  among other reasons because the high degree of structural homogeneity makes these materials appropriate to investigate the influence of disorder in ideal periodic vortex lattices. The weak collective pinning concept introduced by Larkin and Ovchinnikov<sup>4</sup> to predict the behavior of superconducting vortices is also used<sup>5</sup> to discuss the properties of other periodic systems. Luzuriaga<sup>6</sup> recently remarked that the understanding of pinning interactions can provide new information concerning the nature and density of defects in disordered matter.

In this work we are concerned with the behavior of very dilute vortices. We define the elementary interaction as that between a single vortex and defect distribution.

When studying this elementary pinning interaction in  $Zr_{70}Cu_{30}$  we have found that vortex pinning induces an anomalous increase in the magnetic flux through the sample, when decreasing temperature at constant field. The effect is explained assuming that the number of vortices in the sample is conserved and taking into account the magnetic interaction between the vortices and the surface of the sample. The temperature dependence of this flux expulsion is used to determine the average position where the vortices are pinned.

Pinning forces in superconducting amorphous metals can become very weak,  $1^{-3}$  indicating a high degree of homogeneity at the scale of the superconducting coherence length,  $\xi(T)$ , typically 100 Å.

In an ideal infinite sample the density of vortices is unequivocally determined by the magnetic field H and temperature T. In this case the magnetic flux through the sample should vary reversibly with T and H.

A distinctive property<sup>7</sup> of vortex pinning is the irreversible superconducting magnetization associated to the mixed state. Nevertheless, it is necessary to remark that although the irreversibility is caused by the presence of pinning centers, it can only be detected when the force on the vortex system is large enough to modify the density of vortices in the sample. Our results were obtained studying the behavior of the magnetic flux through amorphous samples as a function of temperature in the very dilute vortex regime.

To be able to detect the flux associated with a single vortex we use a superconducting quantum interference device (SQUID) magnetometer setup<sup>8</sup> that allows the measurement of flux changes as a function of temperature, while the field is kept constant. The overall sensitivity is half a flux quantum.

The Zr<sub>70</sub>Cu<sub>30</sub> samples are melt-spun ribbons, 1.5 cm long, 0.04 cm in width, and a thickness of 14  $\mu$ m. Their superconducting and normal properties as well as the thermal relaxation effects have been thoroughly studied in our laboratory.<sup>2,8</sup> The sample is mounted on an oxygen-free copper support weakly coupled to a <sup>4</sup>He evaporator. The <sup>4</sup>He evaporator is maintained at 1.4 K and the temperature is swept using an electric heater. The sample temperature is measured with a calibrated carbon thermometer. Spurious magnetic signals are within the experimental noise. The temperature sweep and the SQUID are automatically controlled by an Apple II computer. The sample was found to be in excellent thermal contact with the sample holder, within the time scales of our experiment. The magnetic field was applied by means of a superconducting magnet operating in the persistent mode. It is parallel to the surface of the sample within 2°. The remanent magnetic field was reduced to 30 mOe by a proper shielding.

The measurements were performed by cooling the sample down to 1.4 K at zero applied field, then turning on the magnetic field. The temperature was increased and the change in the magnetic flux was recorded. If the field and the temperature were kept low enough, the sample had no vortices. In this case the sample is in the Meissner state and the reversible magnetic flux change, detected by the SQUID, is induced by<sup>8</sup> the temperature variation of the penetration depth,  $\lambda(T)$ .

The vortex penetration into the sample is detected either by the deviation from the BCS temperature dependence of the Meissner magnetization or by the irreversible temperature dependent behavior of the flux expulsion.

The experimental results discussed in this work are obtained at temperatures and fields where the distance among vortices are much larger than  $\xi(T)$ . In this limit, the magnetic flux through the sample is due to the linear superposition of the fields induced by the vortex currents plus the external field penetrating through the sample surface. Under this assumption the magnetic flux associated with the vortex distribution is the difference between the measured flux expulsion and that due to the Meissner state. The Meissner flux was obtained measuring the flux change as a function of temperature at very low fields. The experimental data for fields up to 0.5 Oe are extremely well fitted by the BCS theory (with size effects incorporated) up to T = 2.63 K. This is the temperature chosen as  $T_c$  of the sample. This value is in agreement with the experimental flux expulsion that shows no variation of flux at temperatures higher than 2.65 K. In the measurements, the magnetic field was chosen low enough to assure that at the lowest temperatures, and for the first sweep, the sample remained in the Meissner state. This was verified checking that the expulsion of flux from the lowest temperatures up to the critical temperature,  $T_c$ , scaled with the applied field.

Figure 1 shows typical results for the temperature dependence of the vortex flux, defined as the difference between the total flux and the Meissner flux. In this particular case the data corresponds to a field of 3.5 Oe. It is seen that at the beginning of the first run the vortex flux remains zero. At a well-defined temperature,  $T_{c1}$ , a sudden change in the temperature dependence of the magnetic flux indicates the vortex penetration. Further increase in temperature is followed by a continuous flux increase. Before reaching  $T_c$ , when the temperature is decreased the flux increases up to a maximum and then decreases either to zero or to a finite value, if vortices are trapped within the sample. The temperature is increased again up to a higher value than that of the previous loop repeating the procedure several times for each applied field. For clarity only some temperature runs are shown in Fig. 1.

Several features related to the data shown in Fig. 1 should be noticed. The zero-flux region in the first run indicates the temperature range where the sample remains in the Meissner state. Since the flux sensitivity detection is half flux quantum, the continuous increase of flux above  $T_{c1}$  indicates that the vortices enter without flux jumps. The hysteretic behavior of the magnetic flux and the flux trapped at low temperatures are clear indicators of flux-pinning forces. As a consequence, we expected the magnetic flux to remain constant when decreasing temperatures.



FIG. 1. Temperature dependence of the vortex flux, defined as the difference between the total flux and the Meissner flux. Applied field is 3.5 Oe. Arrows indicate the direction of the temperature sweeps. Anomalous reversible regions are indicated by solid circles. For clarity only some temperature runs are shown.

ature, until the state with a fixed number of vortices becomes energetically unstable. It was surprising to find that the magnetic flux increases reversibly before the nonreversible decrease was detected, as indicated in Fig. 1.

In what follows we show that the reversible flux change is consistent with a finite superconducting system that sustains a constant number of pinned vortices. To understand the phenomenon it is sufficient to consider the behavior of a single vortex with its core pinned at a distance  $x_s$  from the surface of the sample. If  $x_s \gg \lambda(T)$ , the flux of the vortex should be quantized and its contribution to the sample flux would be just one flux quantum,  $\Phi_0 = 2 \times 10^{-7}$  Oe cm<sup>2</sup>. On the other hand, if  $x_s \simeq \lambda(T)$  the fluxoid is quantized and the magnetic flux contribution depends on  $x_s / \lambda(T)$ , the effective distance in this problem. When the temperature is decreased, the effective distance increases raising the contribution of the vortex to the flux through the sample, as observed experimentally.

Let us express this behavior in a more formal way. The superconducting penetration depth in this material has been found<sup>8</sup> to be  $\lambda(0) \simeq 1 \mu m$ . Consequently, the high- $\kappa$  approximation ( $\kappa = \lambda/\xi$ ) is well justified when studying the electromagnetic response of the superconductor.

The magnetic induction field **h** within a high- $\kappa$  superconductor containing a single vortex can be approximated by the solution of the equation<sup>9</sup>

$$\lambda^2(T)\nabla^2 \mathbf{h} - \mathbf{h} = -\Phi_0 \delta^2(x, y) \hat{\mathbf{z}} , \qquad (1)$$

where  $\hat{z}$  is a unit vector along the vortex,  $\delta^2$  is a twodimensional delta function, and the core of the vortex is assumed to be at x = y = 0. If the sample is infinite the solution of Eq. (1) is  $\mathbf{h}(r) \sim K_0(r/\lambda)\hat{\mathbf{z}}$ , where  $K_0$  is the modified Bessel function of zero order. The magnetic flux through the sample is  $\Phi_0$ . If the sample containing the vortex is considered to be semi-infinite with an external magnetic field H applied parallel to the surface, Eq. (1) should be solved with appropriate boundary conditions. The field h should be parallel to H and is due to the superposition of the surface field  $\mathbf{h}_s$  and the vortex field  $\mathbf{h}_v$ . The surface field is the solution of the homogeneous London equation  $\lambda^2(T)\nabla^2 \mathbf{h}_s - \mathbf{h}_s = \mathbf{0}$  with the boundary condition  $\mathbf{h}_s = \mathbf{H}$  at the surface. The vortex field should satisfy Eq. (1) with the boundary condition  $\mathbf{h}_v = \mathbf{0}$  at the sample surface. Let us consider the sample filling the space from  $x = -\infty$  to  $x = x_s$  and the vortex with the core at x = y = 0. Making  $\mathbf{h} = \mathbf{h}_v$  in Eq. (1) and integrating over the cross section of the sample perpendicular to the z axis, and taking into account that  $\mathbf{h}_v = 0$ at distances from the core much larger than  $\lambda(T)$ , we obtain

$$\lambda^{2}(T) \frac{d^{2} \Phi(x_{s})}{dx_{s}^{2}} - \Phi(x_{s}) = -\Phi_{0} , \qquad (2)$$

where  $\Phi(x_s)$  is the magnetic flux defined by  $\Phi(x_s) = \int_{-\infty}^{+\infty} dy \int_{-\infty}^{x_s} dx h_v(x,y)$ . Solving Eq. (2) with the condition  $\Phi(x_s) \rightarrow \Phi_0$  when  $x_s \rightarrow \infty$  we find

$$\Phi(x_s) = \Phi_0(1 - e^{-x_s/\lambda}) .$$
(3)

As discussed previously, this equation contains the

main features observed in the experimental data shown in Fig. 1. If the vortex is pinned its flux will increase when  $\lambda(T)$  diminishes with temperature. It also shows that the flux of the vortex reduces to zero at  $T = T_c$ .

Consider now a slab of thickness d with the vortex parallel to the surface of the sample. The core is supposed to be at a distance  $x_0$  from the center of the slab, taken at x = 0. Equation (1) may now be solved by means of the method of images. The field is obtained by the superposition of an infinite series of vortices and antivortices which satisfies the required boundary conditions. The vortex flux may be calculated pairing the images equidistant from each surface and using (3). The result is

$$\Phi(d, x_0) = \Phi_0 \left[ 1 - (e^{x_0/\lambda} + e^{-x_0/\lambda})e^{-d/2\lambda} \\ \times \sum_{n=0}^{\infty} (-1)^n e^{-n(d/\lambda)} \right].$$
(4)

This geometrical series leads to

$$\Phi(d, x_0) = \Phi_0 \left[ 1 - \frac{\cosh(x_0/\lambda)}{\cosh(d/2\lambda)} \right] .$$
(5)

If there were more than one vortex in the sample and if the distance between vortices is much larger than  $\xi(T)$  the total flux would be

$$\Phi_T = \Phi_0 \sum_{i=1}^{N} \left[ 1 - \frac{\cosh(x_i/\lambda)}{\cosh(d/2\lambda)} \right], \tag{6}$$

where N is the number of vortices and  $x_i$  is the position of the *i*th vortex.

If we assume that all vortices are pinned at a welldefined distance  $x_0$  from the center, expression (6) is reduced to

$$\Phi_T(d, x_0) = N \Phi_0 \left[ 1 - \frac{\cosh(x_0/\lambda)}{\cosh(d/2\lambda)} \right] . \tag{7}$$



FIG. 2. Universal behavior of the vortex flux as a function of temperature. Applied field is 3.5 Oe. Normalization factors are inversely proportional to the number of pinned vortices at each temperature sweep (see text). Solid lines are fits from Eq. (7), with the following parameters. Upper curve: N = 930,  $x_0 = 4\lambda(0)$ ; middle curve: N = 780,  $x_0 = 3\lambda(0)$ ; lower curve: N = 700,  $x_0 = 2\lambda(0)$ .



FIG. 3. Same as in Fig. 2, with an applied field of 5.3 Oe. Fit parameters are the following. Upper curve: N = 1280,  $x_0 = 3\lambda(0)$ ; middle curve: N = 1150,  $x_0 = 2\lambda(0)$ ; lower curve: N = 1080,  $x_0 = \lambda(0)$ .

In this case the vortex flux normalized by the number of vortices is only a function of the reduced distance  $x_0/\lambda(T)$ .

Expression (7) suggests a convenient way of representing the experimental data to determine if the vortices are distributed around a well-defined average value  $x_0$ . If that were the case, flux change should have the same temperature dependence for all the temperature sweeps at the same applied field. Different curves would differ only in a scale factor N. Since this number of vortices is not known, we plot the vortex flux as a function of temperature for one of the curves, then we try to overlap the other curves corresponding to the same field, multiplying the flux in each one by a different constant. If a universal behavior is found, the normalization factors correspond to the ratio between the number of vortices locked in the reference curve, to that of the curve under consideration. The procedure is repeated for each applied magnetic field. Figures 2-4 show the result for this type of plot. The curves that correspond to consecutive temperature sweeps are identified by consecutive letters. The flux scales in the



FIG. 4. Same as in Fig. 2, with an applied field of 9.6 Oe. Fit parameters are the following. Upper curve: N = 2410,  $x_0 = 2.5\lambda(0)$ ; middle curve: N = 2220,  $x_0 = 1.5\lambda(0)$ ; lower curve: N = 2130,  $x_0 = 0.5\lambda(0)$ .

TABLE I. Average distance of the vortices from the center of the sample, measured in units of  $\lambda(0)$ , for different applied fields. Thickness of the sample is  $14\lambda(0) \simeq 14 \ \mu m$ .

H (Oe)	$x_0/\lambda(0)$	
1.7	3±1	
3.5	3±0.5	
4.4	$2.5 {\pm} 0.5$	
5.3	2±0.5	
7.1	1±0.5	
9.6	$1.5 {\pm} 0.5$	

figures correspond to the last temperature sweep at each field (curves D, F, and E in Figs. 2, 3, and 4, respectively). The other curves are multiplied by the indicated normalization factor.

We see that there is a range of temperatures where the experimental data follow a universal behavior extremely well. The deviation from the universal behavior occurs at a well-defined temperature, where vortices start to leave the sample. The universality for a given field and different number of vortices indicates that, within the experimental accuracy, the vortex distribution in the sample is independent of temperature. On the other hand, the curves are not universal with regard to the magnetic field dependence, indicating that the vortex distribution in the sample is influenced by the external magnetic field.

If universality is due to a narrow distribution of vortices centered at  $x_0$ , expression (7) should fit the experimental universal curve. The solid lines in Figs. 2-4 indicate the result of the fit for different values of N and  $x_0$ , using  $\lambda(T)$  obtained from the data at fields lower than 0.5 Oe. These pairs of parameters are chosen to fit the experimental data near  $T_c$ . The difference among the curves at lower temperatures indicate the sensitivity to the variation of those parameters. The fits in the figures correspond to the following parameters  $[x_0]$  is expressed in units of  $\lambda(0)$ ]. Figure 2: upper curve, N = 930,  $x_0 = 4\lambda(0)$ ; middle curve, N = 780,  $x_0 = 3\lambda(0)$ ; lower curve, N = 700,  $x_0 = 2\lambda(0)$ . Figure 3: upper curve, N = 1280,  $x_0 = 3\lambda(0)$ ; middle curve, N = 1150,  $x_0 = 2\lambda(0)$ ; lower curve,  $N = 1080, x_0 = \lambda(0)$ . Figure 4: upper curve, N = 2410, $x_0 = 2.5\lambda(0)$ ; middle curve, N = 2220,  $x_0 = 1.5\lambda(0)$ ; lower curve, N = 2130,  $x_0 = 0.5\lambda(0)$ . The results for  $x_0$  obtained from the best fitting can be seen in Table I.

Although this result cannot be taken as a proof, the reasonable numbers in Table I and the excellent fitting in Figs. 2–4 strongly indicate that the position of the vortices in the sample is mainly determined by the force exerted by the external magnetic field and a uniform distribution of pinning centers. The vortex-vortex interaction does not seem to play a fundamental role in the range of temperatures where the universality is obeyed. This is also supported by the systematic shift of  $x_0$  towards the center of the sample when H is increased.

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Another explanation based only on equilibrium properties of finite samples could be considered: In this case the vortices would remain at fixed positions when changing temperature, due to the vortex-vortex repulsion maintaining the vortices against the potential surface barrier.<sup>10</sup> An ideal barrier should preclude<sup>11</sup> vortex penetration up to a temperature where the thermodynamic critical field equals the applied field. After the vortices have penetrated into the sample, they should remain trapped down to zero temperature because the average internal field B is lower than the applied field H. This behavior is not supported by the experimental data shown in Fig. 1. Even for very diluted vortex concentrations, the vortices start to leave the sample at the same temperature where they penetrated indicating that the surface barrier is almost suppressed in certain regions of the sample. On the other hand, the smooth flux increase after the first vortex enters into the sample is a strong indication of the existence of bulk pinning. A sample of macroscopic dimensions free of inhomogeneities should show<sup>12</sup> an almost infinite slope of  $\Phi(T)$  at  $T_{c1}$ .

It is interesting to point out that if the sample has an ideal surface barrier and no bulk pinning, the theory<sup>10</sup> predicts that there is a region near the surface of the sample, of thickness  $\lambda(T)\cosh^{-1}(H/B)$ , free of vortices. As a consequence, if T decreases at a constant applied field, the vortices move towards the surface. This is in contradiction with the assumption made to explain the flux anomaly.

In this paper we have shown that the magnetic flux through finite samples, associated to a fixed number of vortices, changes reversibly with temperature because the flux associated with a vortex is less than a flux quantum.

The interpretation of the irreversible flux expulsion behavior, as well as the study of the temperatures and fields where the irreversible flux motion takes place, are not the subject of this paper, but the experimental data already show that the technique used to investigate the magnetic response of diluted vortices is useful for the investigation of the elementary pinning forces.

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