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Anomalous flux pinning in a torus of thoriated UBe_{13}

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We compare the local magnetization at a spatial singularity of superconducting $U_{0.97}Th_{0.03}Be_{13}$ for different magnetic field histories. We find a reduction in the zero-field-cooled vortex flow relative to the field-cooled case, marked by a sharp onset coincident in temperature with the second (lower-temperature) transition in the specific heat. This effect is peculiar to the flux threading the hole of the torus, to (U,Th)Be_{13} samples with multiple superconducting transitions, and to the lower superconducting state with broken time-reversal invariance.

The heavy-fermion system (U,Th)Be_{13} has an unusual phase diagram with several superconducting phases. For a certain range of Th concentrations, there exist phase transitions within the superconducting state as functions of either temperature[1] or pressure.[2] These transitions could be purely magnetic in nature or, as the weight of the evidence suggests, they could stem from a fundamental change in the superconductivity.[3] The existence of multiple superconducting phases is strong evidence of unconventional superconductivity. In this case, the important problems arise of finding the order parameters that describe the different phases and determining why the free energies are so close that more than one phase is observed.

There are relatively few experimental probes for investigating the multiple transitions. Once a sample is superconducting, measurements such as resistance and susceptibility do not detect additional phase boundaries. Even the phase boundary in temperature, the easiest to measure, has been seen with only a few techniques. For Th concentrations between 1.8 and 4.5 %, specific-heat measurements[1] show a transition at temperature T^* in addition to the initial superconducting transition at $T_c > T^*$. The lower critical field, H_{c1}, is linear in T^2 but has an abrupt change in slope[4,5] at T^*. The second transition is marked as well by a large peak in the ultrasonic attenuation.[6] Finally, for $T < T^*$, muon spin relaxation (μSR) measurements[7] reveal a local magnetic field which probably originates from a superconducting phase with broken time-reversal symmetry. Other regions of the temperature-concentration phase diagram do not provide evidence for a local field. Apart from the μSR data, no experiments thus far have restricted the form of the order parameter in any of the phases.

A nonscalar order parameter can lead to particularly unusual behavior at a spatial singularity. In the ^{3}He superfluids, which have spin triplet pairing, both free vortices and circulation around a physical obstruction possess interesting properties. For example, both ^{3}He-*A* and ^{3}He-*B* exhibit phase transitions in their free vortices.[8] Furthermore, ^{3}He-*B* circulation can be trapped[9,10] partly or fully around a spatial singularity, while bulk ^{3}He-*A* does not support persistent currents in a toroidal geometry.[9] In the measurements reported here, we extend the ^{3}He analogy to thoriated UBe_{13} by using a physical hole in the sample as a pinning center for vortices. Flux trapped in a hole behaves in many ways like an individual vortex in the bulk, but the large quantum numbers allowed and the enforced immobility of the core may alter other aspects of the response.

We measured the local magnetization of high-purity, long-term-annealed polycrystals[11] of $U_{0.97}Th_{0.03}Be_{13}$, and of pure UBe_{13} for the purpose of comparison. The thoriated material has two clear transitions in temperature, while the pure substance has only one. Sample shapes were irregular, with 0.7 mm thickness and planar dimensions ranging from 1.5 to 6 mm. We drilled holes approximately 0.6 mm in diameter using a spark cutter with a 23 gauge copper wire as the cutting element. A small solenoid generated an external field $H \leq 100$ Oe along the axis of the hole. We measured the local field B with a bismuth Hall probe of active area 3 μm$\times$5 μm.[12] The gaussmeter was glued to the sample with GE 7031 varnish and it could be moved upon thermal cycling to determine the magnetization either through the hole or in the bulk. The toroidal sample, the Hall probe, and the magnet (as sketched in the inset to Fig. 1) were cooled to temperatures between 0.3 and 1 K on a charcoal-pumped ^{3}He cryostat.

Both theory and experiment[7] suggest that the superconducting state below T^* violates time-reversal symmetry and, consequently, possesses various unusual magnetic properties. In particular, the interaction of the lower temperature phase with a magnetic field may differ from that of a superconductor where time-reversal invariance is conserved. For example, the local field seen in the lower phase may be influenced by a finite H on cooling below T^*. With this expectation in mind, we measured both the zero-field-cooled (ZFC) and the field-cooled (FC) magnetization. Our ZFC procedure involved cooling the sample from the normal state ($T > 0.65$ K for the thoriated samples, $T > 0.95$ K for the pure

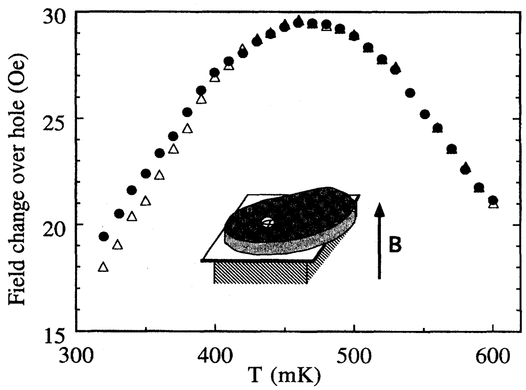

FIG. 1. Difference between magnetic field through the hole in a torus of $U_{0.97}Th_{0.03}Be_{13}$ at maximum applied field $H_m = 20$ Oe and at zero field. Curves follow zero-field-cooled (△)and field-cooled (●) protocols, as described in the text. Illustration shows a sample with its hole over the active region of the Bi Hall probe, linked to the cryostat by a copper rod.

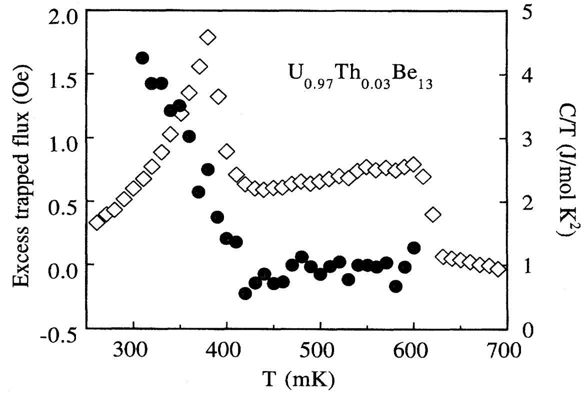

FIG. 2. Specific heat C (◇, right axis) and excess trapped flux (●, left axis) vs temperature T for $U_{0.97}Th_{0.03}Be_{13}$. The onset of excess flux trapping for the ZFC procedure coincides with the lower temperature jump in C/T.

material) to a temperature in the superconducting state and stable to within 0.001 K. We applied a maximum magnetic field H_m and measured the local magnetization of the sample, then reduced the applied field to zero and measured the magnetization again. The values we find repeat on subsequent measurements at the same maximum field H_m and at $H=0$.

On field cooling, we applied the field H_m in the normal state before cooling. We observed little Meissner effect; that is, the initial field on cooling into the superconductor is very close to the applied field, rather than close to zero because of flux expulsion. This result is consistent with previous observations[5] of Meissner fractions near 25% at very low fields, but a saturation in the amount of flux expelled by applied fields of a few gauss. We measured the trapped field after removing the external field, and the magnetization on returning to the field H_m. As for the ZFC case, these values repeat on further cycling of the field. All comparisons between ZFC and FC data use the same field $6 \leq H_m \leq 80$ Oe as the maximum applied field and as the field for cooling in the FC run.

After zero-field cooling, the initial magnetization curve should show $B=0$ until the applied field reaches H_{c1}. From measurements over the bulk and in samples without a hole, both pure and thoriated UBe_{13} show $B>0$ by $H=1$ Oe. Previous authors have found H_{c1} of 10 to 30 Oe at our temperatures.[4,5] We may see no H_{c1} because our samples have irregular surfaces which can cause large demagnetization effects, pushing the field for flux entrance to very low values, or because they are polycrystals and flux can enter easily along grain boundaries.

We find that ZFC and FC preparations lead to essentially equivalent results both in conventional solid samples and in local regions away from the hole in the toroidal topology. More flux is trapped on FC preparation, probably because some of the large number of vortices initially present pass through crystallites rather than lying completely along grain boundaries. These vortices are more strongly pinned than the interstitial vortices and remain trapped in the sample after the field is removed. However, the *difference* between the magnetization at maximum field H_m and at zero field is unchanged between the FC and ZFC protocols. This difference corresponds to the number of vortices that leave the sample as the field is turned off and probes the forces impeding vortex motion.

By contrast, the amount of vortex motion over a hole in $U_{0.97}Th_{0.03}Be_{13}$ below T^* depends strongly on cooling conditions. We show in Fig. 1 the difference between the magnetization at $H_m = 20$ Oe and at $H=0$ for both ZFC and FC preparations. The two curves agree at high temperatures but deviate near 410 mK, with the vortex motion clearly less in the ZFC case. In order to show the deviation more clearly, we plot in Fig. 2 (left axis) the difference between the FC and ZFC curves, as a function of temperature. We refer to this quantity as the "excess trapped flux." As we discuss below (viz., Fig. 4), the difference arises from the enhanced trapping of the ZFC preparation near $H=0$. Specific-heat data for the same polycrystal, also plotted in Fig. 2 (right axis), demonstrate clearly the agreement between the lower transition temperature T^* and the onset of the reduced ZFC vortex motion.

After first observing this effect, we drilled a second hole in the same thoriated sample. That hole also shows the reduced vortex motion for ZFC, with the same onset temperature. Moreover, no such effect is seen over a hole in pure UBe_{13}, at temperatures down to 320 mK and for $6 \leq H_m \leq 80$ Oe. The coincidence of the onset temperature with T^* and the absence of any effect in the pure sample indicate that the behavior is truly a property of the low-temperature phase in $U_{0.97}Th_{0.03}Be_{13}$.

The difference in trapped flux sensed by the magnetometer between the ZFC and FC preparations reaches a maximum of 2 Oe, corresponding to approximately 25 000 flux quanta threading the hole. Since this field is small, we have considered the possibility that it results from a critical-field phenomenon. In such a scenario, the vortex motion could be reduced in the ZFC case until some critical field is exceeded, but the large amount of trapped flux arising from FC preparation makes this critical field irrelevant. Such a field should be temperature dependent, most likely increasing with decreasing T. Hence, the match between the onset temperature and T^* in our data (Fig. 2) would be only a coincidence, and

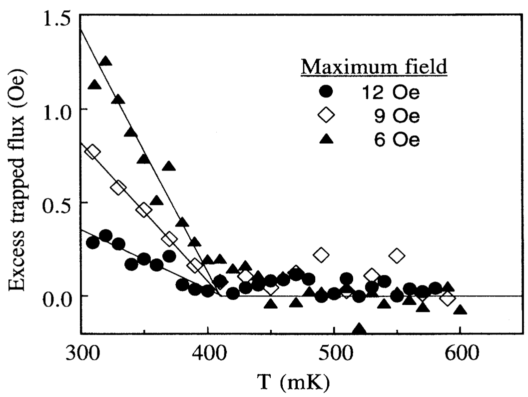

FIG. 3. Excess trapped flux as a function of temperature for three values of maximum field, H_m. The horizontal line is at zero excess flux. Other solid lines are linear fits to the data below 390 mK; all intersect zero at $T=410\pm1$ mK.

the onset temperature should vary with H_m.

We have checked this possibility explicitly, as illustrated in Fig. 3 for maximum applied fields of 6, 9, and 12 Oe. The low-temperature lines are best fits to the data for $T\leqslant390$ mK. Although H_m changes by a factor of 2, all three fits extrapolate to onset temperatures of 410 ± 1 mK. From varying which data points are included in each fit, we estimate an absolute uncertainty of ±5 mK. Assuming that a critical field would scale as $[1-(T/T_c)^2]$, as does H_{c1} for example, we calculate that a change in H_m by a factor of 2 should correspond to a temperature change from 410 of over 100 mK. Furthermore, any field effects should have been apparent in the pure UBe_{13} sample as well, again inconsistent with our observations.

A hysteresis loop for $H_m=7$ Oe with the gaussmeter placed over the hole (the outermost curve in Fig. 4) shows clearly that less vortex motion is seen for ZFC because of increased vortex trapping at low fields. The nearly flat region at external fields less than 1 Oe and the rapid change in the internal field (and the magnetization) near 1 Oe show a strong pinning effect at very low fields. This structure is seen only for ZFC. On field cooling, the low-field hysteresis disappears. The behavior is also temperature dependent, with the width narrowing as temperature increases. Above T^*, no feature remains.

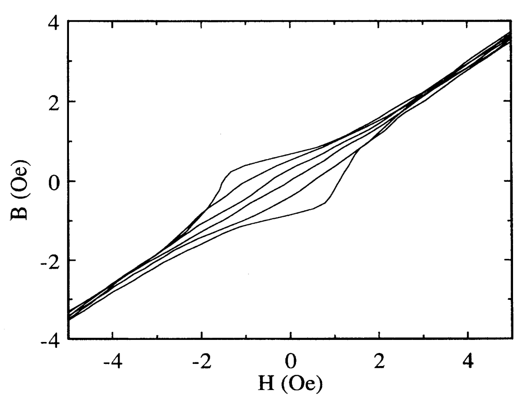

FIG. 4. Low-field portions of hysteresis loops at $T=320$ mK after ZFC preparation. The maximum fields were 7, 12, and 15 Oe (from outside to inside).

Vortices threading another torus of $U_{0.97}Th_{0.03}Be_{13}$ needed to surmount a surface barrier to flux penetration of 12 Oe, giving the hysteresis loop a different shape and substantial width. However, subtracting FC from ZFC hysteresis loops reveals a sharp increase in magnetization at low fields over this hole as well. The data presented on vortex motion in Figs. 2 and 3 are from two different holes, but display essentially the same FC-ZFC difference.

The nested hysteresis loops of Fig. 4 have a peculiar dependence on H_m. As the maximum applied field is increased, the low-field bulge narrows, but the high-field response is not affected. We concentrate in Fig. 4 on the low-field behavior after maximum applied fields of 7, 12, and 15 Oe. At fields above 2.5 Oe, the three loops coincide within our measurement precision. As H is decreased, the three curves diverge. The variation with H_m seen here underlies the reduced ZFC vortex motion dependence on H_m of Fig. 3. We have checked that magnetization over the bulk indeed is independent of H_m.

The critical role played by the ZFC state may be a manifestation of broken time-reversal invariance below T^*. One possible explanation for our observations is that the local magnetic field of the lower superconducting phase forms magnetic domains. Domain walls act as pinning centers for vortices, which can decay into fractional vortices (i.e., line defects in the walls which are vortices enclosing a fraction of the universal flux quantum).[3,13,14] The ability of the walls to pin vortices is enhanced because fractional vortices cannot exist off the wall and must recombine before leaving. A domain wall with many trapped vortices is a barrier to the motion of other vortices through the repulsive vortex-vortex interaction. The highly reduced flux creep recently observed in the low-temperature phase of the heavy-fermion superconductor UPt_3 has been attributed to the same barrier mechanism.[15] For UPt_3 there are also indications that the low-temperature superconducting phase breaks time-reversal symmetry.

In our experiment, the ZFC protocol leads to a random distribution of domain sizes and alignments, and consequently many domain walls. The FC counterpart, resulting in many trapped vortices, may "polarize" the superconducting order parameter and have a preferred domain type. The FC domain alignment may persist after the field is removed because the vortices trapped within grains prevent the local field from reversing. Hence, a FC history can lead to fewer domain walls than a ZFC procedure, and to a correspondingly larger flux flow.

We only detect a difference between ZFC and FC procedures when the magnetometer is placed over the hole. This observation may be explained by the fact that the ability of domain walls to block vortex motion can be particularly high at the surface of the sample, where entering vortices quickly can saturate the available space. By contrast, even a few walls crossing a large sample could enclose most of the vortices, making vortex flow through the bulk less dependent on the density of domain walls.

In summary, we have discovered a method of observing the lower transition in $U_{0.97}Th_{0.03}Be_{13}$ by employing a toroidal topology and a magnetometer of micron dimensions. The freedom of vortices to move from the bulk of a sample into a hole depends, in the lower phase only, on whether or not the sample was cooled in a magnetic field. The zero-field-cooled preparation leads to excess flux trapping at low fields, best seen in the unusual shape of the hysteresis loop. Our experimental findings may be linked to the interactions of the vortices with the local magnetic fields which characterize the lower superconducting state.

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