An investigation of the appearance of positive magnetic moments on field cooling some superconductors

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"Paramagnetic" moments from some high-Tc superconductors, measured using a SQUID magnetometer, are presented. Similar results have been published elsewhere (W.M. Lee et al., Solid State Commun. 74 (1990) 97.) with attempts to explain them in terms of a material property. We have investigated this phenomenon thoroughly and we show here adequate evidence that these moments are an artifice, a combination of peculiar circumstances in the machine. They arise largely through the flux trapping capabilities of the material and the inhomogeneity of the magnet in the machine. They can be explained using the known behaviour of materials containing trapped flux. They are not due to the remanent field of the magnet.

1. Introduction

In many laboratories it is common practice to characterise a new superconductor in the following way:
(a) oscillate the field in the magnetometer to 5 T,
(b) oscillate the field to zero (this ensures that the lowest possible remanent field is left in the machine),
(c) zero field cool the sample in the magnetometer,
(d) apply a small field (usually less than 100 G, sometimes less than 10 G),
(e) warm the sample and monitor its magnetic moment,
(f) cool the sample in the field and monitor its magnetic moment.

Step (f) is widely believed to give the fraction of superconducting material present, but should be interpreted more in terms of flux pinning. Striking evidence of this is that zero field cooled and field cooled experiments on pure lead give only a relatively small "Meissner fraction" - typically 50%. In addition field cooled experiments on strongly pinning samples, such as melt texture samples, give very low "Meissner fractions". A detailed discussion of this is given in various publications of Kitazawa et al. [2,3]. The zero field cooled moment gives some indication of quantity of superconductor present. This is discussed in some detail in another paper [4].

Leaving the interpretation of the Meissner fraction aside, there are a number of questions that should be addressed in making this measurement. Is the specimen really being cooled in zero field, and does this set a lower practical limit on the applied field that can be used? How uniform is the magnetic field? When the machine warms and cools the specimen is the temperature rise and fall monotonic?

Our attention was drawn to a set of samples which had a diamagnetic moment when subjected to procedure (e) above, but a positive moment when subjected to procedure (f). Results similar to this have been published [1], citing paramagnetic inclusions as a possible source of the moment. However, the magnitude of the positive moment shows some peculiarities. In particular the moment reported was larger as the field was reduced, and appeared to begin at a temperature below $T_c$ of the material. This suggested that paramagnetic inclusions were an unlikely cause.

We undertook a systematic series of measurements in order to elucidate the mechanism by which the positive moment was produced.
2. Experimental

The majority of our measurements were carried out on a quantum design SQUID magnetometer, equipped with a 5.5 T magnet. As will be seen, careful consideration of the manufacturer's literature, and keeping a watchful eye on the machine while the measurements are being made would have warned the user that the results were spurious. The manufacturer provides the machine with the facility to monitor the sample response function continuously, and the temperature is displayed at frequent intervals. The magnetometer assumes that the sample is a point source. All corrections due to the true physical shape of the sample must be made by the user. A certain background signal is produced by the sample holder. This was in general two to three orders of magnitude smaller than the signal coming from the sample in this investigation, and was relatively insensitive to temperature in this temperature range.

The superconducting sample used in this investigation was \((\text{Tl}_{0.5}\text{V}_{0.5})\text{Sr}_2(\text{Ca}_{0.8}\text{Y}_{0.2})\text{Cu}_2\text{O}_y\) in the form of a small sintered piece, which approximated to a cylinder 1 mm diameter. The AC susceptibility of this sample is shown in fig. 1. The field in the susceptometer is 10 µT. The susceptometer trace shows the behaviour typical of high-\(T_c\) superconductors, with an initial onset of diamagnetism at approximately 100 K, followed by the rapid increase at about 82 K which is probably due to intergrain connections becoming superconducting. We do not have sufficient knowledge about this material to interpret the signals fully. There may be more than one superconducting phase present, but the large change in signal at around 82 K disappears when the sample is ground to a powder.

The positive field cooled moment was produced using the sequence described in the introduction. The field applied at stage (d) was 10 G, and the temperature was swept from 5 to 130 K and back to 5 K. The positive moment was entirely reproducible, and would appear for any applied field below approximately 40 G. The positive moment was, in general, larger as the applied field was diminished. The magnetometer measures the moment by pulling the sample through a set of coils. The scan length was chosen to be 6 cm. This is very significant, and will be returned to later. The results of this measurement are shown in fig. 2. Notice that the onset of the positive moment is around the temperature at which the intergrain connections become superconducting. Using a 2 cm scan length produced negative moments at all times. At a chosen field of 80 G the moments were negative both on zero field cooling and field cooling. This is shown in fig. 3. In principle, measurements using a 6 cm scan or a 2 cm scan should be identical. They are generally not the same for two reasons. The 2 cm scan extracts a smaller proportion of the response function, and the recommended algorithm tends to underestimate the moment slightly when compared to the 6 cm scan. It is also more erratic, since the positioning of the sample is more critical. However, a larger effect in most samples can be

![Fig. 1. AC susceptibility of the sample.](image1)

![Fig. 2. Zero field and field cooled measurements at 10 G, showing the positive moment.](image2)
attributed to slight changes in orientation from one day to another, and also in some cases to the physical deterioration of the sample.

Our first approach was to measure the remanent field, to check that an applied field of 10 G was indeed positive. The remanent field of the superconducting magnet is very history dependent, and if the magnet has been used in its “no overshoot” mode it is negative, and frequently greater than 10 G in magnitude. However, when the magnet has been cycled to a high field in oscillate mode, and cycled down again the remanent field is very small (of order 3 G or less) and negative. It is not possible to measure the remanent field of the magnet with a sample of type I superconductor, as a separate experiment showed that pure lead was capable of trapping up to 50% of the field when it was field cooled [2].

A Hall probe showed that the absolute magnitude of the remanent field was less than 3 G, but that it was not uniform. Using a simple copper coil mounted perpendicular to the axis of the magnet, and an integrator, we mapped out the field pattern under a number of conditions. The coil we used was 1 mm thick, to give good resolution of field with distance, and was calibrated using a known uniform solenoid. The field resolution obtainable was better than 0.1 G, but it was limited in the change that it could measure by the saturation of the integrator. This limited it to around 1000 G. The other limitation was that it was only stable in a uniform temperature environment, and the integrator drifted very slightly. This made it difficulty to measure absolute magnitudes but it was ideal for measuring magnetic field profiles. Figure 4 shows the profile of the remanent field. The absolute magnitude of the field was certainly less than 3 G, but it is most interesting to note that the profile has a distinct minimum in the centre of the magnet. Figure 5 shows the profile measured at 10 G. The minimum is still present, and its depth is of order 1 G. The distance between the two peaks is approximately 7 cm. This local minimum is still present at higher fields, where its presence can affect the measurement of hysteresis in materials which have
very small hysteresis. It is worth noting that a 6 cm scan takes a sample through almost the maximum variation in the central region of the magnet, whereas a 2 cm scan gives the sample a very small variation in field. If the field at the outer edge is 10 G the sample passes through a field of 9 G during its scan, i.e. the field at the centre is of a lower magnitude than the field at the edges.

Knowing that the magnitude of the remanent field was less than 3 G we could be certain that an applied field of 10 G was certainly positive. It occurred to us that if the positive moment from the sample was a material property, reversing the field (i.e. using -10 G at step (d) above) in the measuring sequence should produce identical results, with the signs reversed. This was not observed, and the results of this are shown in fig. 6. Notice that the magnitude of the zero field cooled moment is slightly larger for -10 G, and this is consistent with a remanent field of order -1 G. The measurement of the magnet profile at -10 G is shown in fig. 7. This is quite different from the magnet profile seen at +10 G. Notice that the field in the centre has a larger magnitude than the field at the outer edges. The field cooled magnetisation of the sample in fig. 6 shows no anomalous change of sign.

Because the change of sign always appeared at around the temperature at which the intergrain connections became superconducting we were misled to believe that the sample could be entering a region of lower field and lower temperature simultaneously during a scan, and flux was being frozen into the sample. Supporting evidence was the fact that the hysteresis loop at this temperature had a depth of exactly 1 G. We checked the temperature profile of the machine very carefully, and temperature variations in the sample space are minimal (less than 0.5 K). This mechanism is definitely not responsible.

Further experiments showed that the sign of the magnetic moment of this sample depended upon its precise location in the magnet when it cooled. If the sample was cooled to 10 K without movement in the centre of the magnet, which is where the field is lowest, it invariably had a negative moment, independent of scan length. However, if it was cooled to 10 K 3 cm away from the centre on either side, where the field is highest, it invariably had a positive moment, also independent of scan length. The change in sign of the magnetic moment is therefore produced by cooling in the region of the instrument where the field is highest. The machine then scans the sample through a lower field region to measure its moment. To test this the sample was cooled to 10 K in a 10 G field in the centre of the magnet. It was then moved 1 cm down to be in a position for a 2 cm scan. The magnetic moment was then measured. The field was decreased by 1 G, and the moment remeasured. The sign of the moment had changed. The field was cycled up and down in small steps, up to 4 G at a constant temperature. The moments measured from the sample lay on a reasonably straight line, and the moment passed from negative to pos-
itive for field reductions of order 0.5 G. The results of the field cycling are shown in fig. 8.

This then is the final explanation of the phenomenon. If the sample is measured in a field which is lower than the field in which it was cooled, it may appear to have a positive moment, but if it is measured in the same field, or a higher field it will have a negative moment. The machine generally rests the sample at the lower end of its scan while it changes the temperature, and this is the means by which the positive moment has been produced. Careful examination of the plots of the SQUID scan do show that there is distortion. The crossover points of the scan are not quite in the right places, and the shape of the scan is not quite right. However, most users will miss this fine detail. Typical plots of the machine scans are shown in fig. 9.

3. Model

When a type II superconductor is field cooled, flux pinning sites in the material will tend to trap magnetic flux. This may lead to a field profile in the material resembling that illustrated in fig. 10(a). The average field inside the material is slightly lower than outside, so the magnetic moment of the material is slightly diamagnetic. However, when the field outside the material is changed a number of things may happen. Provided the external field is still below $H_c$, no further flux penetration will occur, and the magnetic moment will increase linearly as the field outside increases. If the field is reduced no trapped flux will escape, and there will come a time when the av-
average field inside the material exceeds the average field outside the specimen. In these circumstances the magnetic moment of the material will appear to be positive. This is illustrated in fig. 10(b). Again, provided the fields are below $H_{el}$ the moment will be linear with field.

In order for this situation to reverse the sign of the moment measured by the SQUID it requires that more than about 95% of the applied field should be trapped in the material on field cooling. This will ensure that the moment of the sample changes sign relative to the local field in the machine as it approaches the centre of its scan. This topic has been given an excellent treatment by Matsushita et al. [3]. They show that the field cooled susceptibility of a hypothetical sample with $T_{c} = 93$ K, diameter 1 mm in a 10 G field will reach less than 10% of the zero field value of the susceptibility if $J_{c}$ is as high as $10^{6}$ A/cm$^{2}$. The trapped flux depends strongly on the volume of material with good superconducting contacts around which the supercurrents may flow. A powder can trap a much smaller amount of flux, even when $J_{c}$ is high. In addition with the new superconductors the London penetration depth is large, which tends to depress the susceptibility of powdered samples. Therefore flux trapping is expected to be most efficient in bulk samples. This was checked experimentally and found to be true. The average magnetisation of a field cooled melt textured sample 1 mm diameter was only approximately 0.25 G less than the nominal 10 G field applied. A powdered version of the material which began this investigation had an average magnetisation 4 G lower than the nominal 10 G field.

Since $J_{c}$ and $H_{el}$ are both material dependent and temperature dependent it is difficult to lay down any guidelines for predicting when this problem will occur. The characteristics of the machine are another factor. Before ascribing a positive moment to a paramagnetic material the measurement needs to be assessed critically.

4. Conclusions

Field cooled specimens are known to trap flux. In some commercial machines this can cause odd reversal of sign in measured magnetic moments if the trapped field in the sample is very close to the field in the machine. This is particularly likely to happen if the machine has a lower field in its centre than at its outer edges. The warning signs are the appearance of less than ideal response functions in the SQUID output, and peculiar variation of moment with applied field. The problem is more likely to occur in bulk specimens, particularly those treated to increase $J_{c}$.

References