Positive field-cooled susceptibility in high-T_c superconductors

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We confirm the recent finding of a paramagnetic Meissner effect on melt-processed $Bi_2Sr_2CaCu_2O_8$ ceramics: on cooling in fields below 200 mG, the samples show a magnetization that is parallel to the applied field. It is shown that the sum rule for the remanent magnetization remains valid in this field range, yielding an absolute value of the remanent magnetization that exceeds the value of the zero-field-cooled magnetization. This observation is discussed in terms of unconventional vortices and current loops. It is argued that the effect is not due to paramagnetic impurities.

Recently, Braunisch *et al.*[1] published superconducting-quantum-interference-device measurements on the Meissner effect of Bi-based high-temperature superconducting (HTSC) ceramics in the very-low-field region: certain samples exhibit a positive susceptibility on field cooling (FC) in fields below 0.5 G, going together with an anomaly in the low-field microwave power absorption. A positive Meissner fraction has already been observed by a number of groups[1–4] and has tentatively been attributed to various reasons, ranging from vortex-pair fluctuations combined with pinning[2] to spontaneous orbital currents at very small fields[1,5] or to the inhomogeneity of the magnet, i.e., an artefact of the measurement.[3]

We examined the low-field Meissner effect of melt-processed $Bi_2Sr_2CaCu_2O_8$ ceramics and observed a positive FC susceptibility in the very-low-field range, in agreement with Braunisch *et al.*[2] Our measurements were performed with a vibrating sample magnetometer, which is equipped with a superconducting magnet. For measurements in the mG regime, a good compensation of the remanent and the earth field is necessary. The remanent field at the position of the sample depends on the magnetic history. The field component parallel to the solenoid axis was compensated with the superconducting magnet to below 10 mG, the perpendicular component by means of a pair of Helmholtz coils to below 40 mG. This is small enough to detect the positive effect. The sample vibration has an amplitude of 1 mm; the variation of the field parallel to the axis follows from the field gradient to be ± 5 mG at most. We measured the FC magnetization at positive as well as negative applied fields in order to rule out an offset in the field zero.

Figure 1 shows the temperature-dependent FC magnetization of a sample at several fields. The four measurements in the upper part of the figure [1(a) and 1(b)] were performed at ± 50 and ± 100 mG. For these fields, the magnetization is positive on cooling in a positive field and negative on cooling in a negative field. In contrast, the magnetization shows the usual diamagnetic flux expulsion on cooling in a field of ± 1 G [Fig. 1(c)]. The magnetization reverses sign upon reversing the sign of the cooling field, for the "paramagnetic" cases *a* and *b* as well as for the diamagnetic case *c*. The value of M is roughly the same for field cooling at 50 and 100 mG, yielding susceptibilities that differ by a factor of 2: $\chi_{FC} \equiv M_{FC}/H \approx 0.15$ for $H = 50$ mG, while $\chi_{FC} \approx 0.08$ for $H = 100$ mG. In contrast, $\chi_{FC} \approx 0.18$ for $H = 1$ G. The zero-field-cooled (ZFC) magnetization of the sample remains perfectly diamagnetic with $\chi_{ZFC} = -1$ within experimental accuracy over the entire field range we examined.

In the temperature range between 4 and 70 K, the magnetization is constant within the noise. The average magnetization of each FC experiment in this temperature range [thin lines in Figs. 1(a) and 1(b)] as function of the applied field is plotted in Fig. 2. For $|H| > 220$ mG, the magnetization shows the usual diamagnetic behavior; below $|H| = 220$ mG it changes sign and becomes

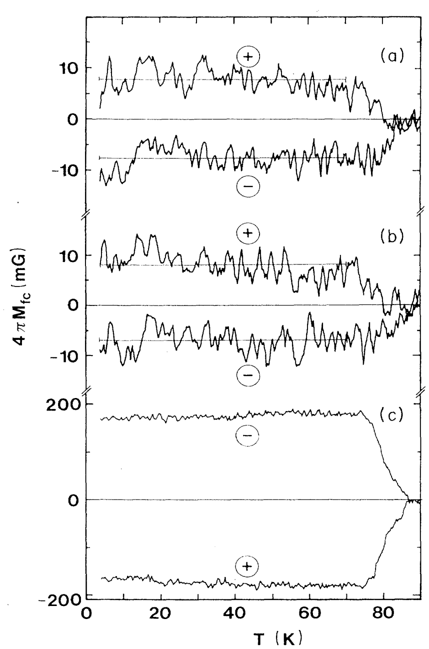

FIG. 1. Temperature-dependent FC magnetization of the sample at ± 50 mG (a), ± 100 mG (b), and ± 1 G (c). The encircled signs indicate the field direction. In the former measurements [(a) and (b)], M_{FC} is parallel to the applied field, while the usual diamagnetic M_{FC} is observed at ± 1 G (c). The average magnetization was determined between 4 and 70 K (thin lines).

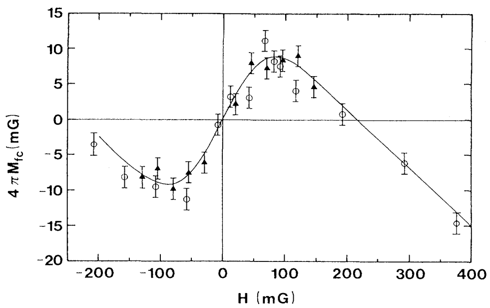

FIG. 2. Field-dependence of the low-field FC magnetization as determined in Fig. 1. Above $|H| \approx 220$ mG, M_{FC} is diamagnetic. Below, the FC susceptibility is positive. M_{FC} reaches a maximum of about 9 mG between 50 and 100 mG. Two sets of measurements are included, where the second (▼) was performed several weeks after the first one (○). The solid line is a guide to the eye.

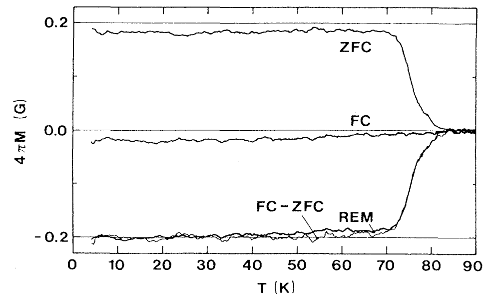

FIG. 3. ZFC and FC magnetization at $H = -107$ mG. The remanent magnetization M_{rem} was measured in zero field after cooling the sample at -107 mG. The thin solid line is the difference of the experimental FC and ZFC line. As it equals M_{rem}, the sum rule for the remanent magnetization is valid.

paramagnetic. The FC magnetization has a maximum of about 9 mG and around 50–100 mG and tends to decrease again towards zero field. Very similar results were obtained in a second set of measurements performed several weeks after the first set, where the perpendicular component of the remanent field was somewhat different (triangles in Fig. 2). In contrast, we did not observe any positive FC susceptibility in a set of measurements, where the perpendicular field was hardly compensated and had a value of more than 250 mG. This field was obviously sufficient to destroy or superpose the effect. This sensitivity to small fields leads to an apparent irreproducibility and is a possible reason why the observation of a positive FC susceptibility has only been published by a few groups to date.

Moreover, we found it important to check whether the positive FC magnetization could be pinned or not. In order to do so, we measured the remanent magnetization of the sample, i.e., the magnetization in zero field while warming up the sample after cooling it down in a small applied field. It turned out that the absolute value of the remanent magnetization after FC below about 200 mG was always larger than the absolute value of the ZFC magnetization at the corresponding field. A precise analysis yielded that the sum rule for the remanent magnetization,[6–9]

$$-M_{rem} = M_{ZFC} - M_{FC} \tag{1}$$

is valid in the low-field range. This is plotted for $H = -107$ mG in Fig. 3: the remanent magnetization equals the calculated difference of FC and ZFC magnetization accurately. This means that the positive FC magnetization is maintained in the sample upon turning the field off as it is conventionally observed for Bi-based HTSC at small fields.[9] Possible consequences of this observation will be discussed below.

We did not observe a positive FC susceptibility on other samples besides the melt-processed ones yet, in contrast to Braunisch *et al.*[1] This may be because of geometrical reasons. The smaller grain size and the weaker intergranular coupling in conventional ceramics possibly lead to an enhanced sensitivity to small fields of the order of our zero field. Melt-processed samples are heated above their melting temperature during processing,[10] and have some features that differ from those of normal ceramics. The density of the sample shown here is about 90% of the x-ray density and is thus much higher than that of normal ceramics.[9–11] The sample hardly has any intergranular regimes, its macroscopic volume is nearly completely shielded up to 250 G. The superconducting transition is rounded off; full (complete) flux expulsion in ZFC measurements is only reached about 10 K below T_c, even at very small fields (Fig. 3). The lower critical field H_{c1} cannot be determined from the intersection of the steep part of the ZFC flux explusion with the constant low-temperature line, as in a recent publication,[11] because strong surface pinning masks the beginning of flux penetration into the sample. This strong pinning is also reflected by the small Meissner fraction $\chi_{FC} \equiv M_{FC}/H$ of the sample, which is below 20% of perfect shielding at most (Fig. 4).

In contrast, the field dependence of the Meissner fraction at higher fields looks quite normal: as is plotted in Fig. 4, it shows a maximum below 1 G. This is the same behavior we observed in a number of $Bi_2Sr_2CaCu_2O_8$ ceramics.[9] There, we interpret the maximum in terms of a crossover from intergranular to intragranular pinning. This interpretation can only be maintained for melt-

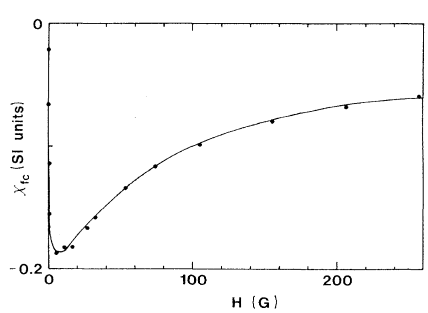

FIG. 4. Field dependence of the Meissner fraction $\chi_{FC} = M_{FC}/H_{loc}$. χ_{FC} shows a maximum at about 10 G and decreases steeply below. It becomes positive below ≈ 220 G (not shown here). The solid line is a guide to the eye.

processed samples that have hardly any intergranular regimes by assuming a granularity of the grains themselves. This is very likely in melt-processed samples, for it is known that their crystalline quality is rather poor, in contrast to the high critical current density, which is due to strong pinning. The assumption of an intragranular cause is also supported by the observation that the positive FC susceptibility is still present after powdering the samples.[1,4]

The reason for the positive FC susceptibility at very small fields is not yet clear and remains a subject of speculation. However, one may discuss possible origins of the effect. First, this is a paramagnetic moment due to the Cu spin or to paramagnetic impurities. This is rather unlikely for several reasons: the susceptibility of Cu(*II*) in the low-temperature range[12] is several orders of magnitude smaller than the susceptibility of the sample measured here, $\chi_{\max} \approx 0.2 = 10$ mG/50 mG (Fig. 2). In order to achieve a sufficient orientation of the Cu spins, a local field has to be assumed that is several orders of magnitude larger than the applied one. This local field would have to occur in the vicinity of T_c, so that the magnetization above T_c remains very small. Moreover, it had to be explained why this paramagnetic magnetization is independent of temperature below T_c but is a function of field as shown in Fig. 2. These arguments hold for paramagnetic impurities as well.

The temperature dependence of the FC magnetization leads to another possibility. M_{FC} remains constant upon cooling, as if the magnetization already reached the saturation value below 1 G and only a few degrees kelvin below T_c, rather pointing to a ferromagnetic magnetization. The remanent magnetization would be due to hysteresis effects then. It can be understood easily in this picture that there is not deviation from the perfectly diamagnetic shielding in ZFC measurements: superconducting surface currents are expected to cancel the field of paramagnetic or ferromagnetic impurities inside the sample.[13] On the other hand, the field dependence of the magnetization remains to be explained, together with the kind of coupling and the value of the Curie temperature.

Thirdly, Braunisch *et al.*[1] use the finding of Bulaevskii, Kuzii, and Sobyanin,[5] to explain the positive FC susceptibility. They argue that for a certain geometry of Josephson loops with magnetic impurities inside the junction, the critical current I_c is negative due to spin-flip scattering in the tunneling barrier. Braunisch *et al.*[1] assume the existence of an average field of interaction between these loops, $H_0 \approx 0.16$ G, below which there is some glassy antiferromagnetic order. They do not argue whether the positive magnetization is expected to be seen in ZFC measurements or measurements of the remanent magnetization as well.

Another possible explanation is given by Svedlindh *et al.*[2], i.e., vortex-pair fluctuations at a Kosterlitz-Thouless transition slightly below T_c. Similarly, Bulaevskii, Ledvij, and Kogan[14] argue that in layered superconductors with moderate anisotropy the spontaneous creation of vortex and antivortex lines should occur above some temperature T_s ($T_s < T_c$). Indeed, the positive magnetization develops only several degrees kelvin below T_c, as was observed previously.[2,4,15] However, since the positive magnetization remains constant upon further cooling, while thermally induced vortex lines are expected only to be visible in the vicinity of the corresponding transition temperature, an additional mechanism has to be assumed to explain the observation. Svedlindh *et al.*[2] suggest that flux pinning plays a crucial role. Consequently, pinning of the paramagnetic vortices would have to be energetically favorable.

In this context, the validity of the sum rule is of some interest because it may yield some information about the nature of the associated currents. The persistence of the paramagnetic currents causing the positive FC magnetization is either due to flux pinning or to the possibility that they are not dependent on field. This possible field independence would be in contrast to the usual diamagnetic surface screening currents which stop flowing upon turning the field off. Such a persistence of the currents cannot be ruled out, but it seems questionable as the FC magnetization decreases with decreasing field in the field range below about 50 mG (Fig. 2). If the persistence of the positive magnetization is due to flux pinning, it must be connected to the existence of vortices. This fits the interpretation of Braunisch *et al.*[1] as well as that of Svedlindh *et al.*,[2] the former assuming some glassy order, the latter flux pinning of Josephson vortices of unconventional origin.

A final decision as to which of the models explains the observations best cannot be made on the basis of our measurements. Further experiments on different samples of well-defined quality are necessary.

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