Equilibrium magnetization in the vicinity of the first order phase transition in the mixed state of high-$T_c$ superconductors.

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We present the results of a scaling analysis of isothermal magnetization $M(H)$ curves measured in the mixed state of high-$T_c$ superconductors in the vicinity of the established first order phase transition. The most surprising result of our analysis is that the difference $\Delta M$ between the magnetization above and below the transition may have either sign, depending on the particular chosen sample. We argue that this observation, based on $M(H)$ data available in the literature, is inconsistent with the interpretation that the well known first order phase transition in the mixed state of high-$T_c$ superconductors always represents the melting transition in the vortex system.

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Measurements of the magnetization of a type-II superconductor in the mixed state provide valuable information about different parameters characterizing the superconducting state of the investigated material. The irreversible magnetization reflects the pinning strength of vortices, and by analyzing the reversible magnetization, different equilibrium parameters of the superconducting material, such as critical magnetic fields and characteristic lengths may be evaluated. This is why magnetization measurements are often used to investigate conventional and unconventional superconductors. For instance, the well-known first-order phase transition in the mixed state of high-$T_c$ superconductors (HTSC), which is usually attributed to the melting of the vortex lattice, was discovered and confirmed by magnetization measurements.\footnote{In this work we apply a recently developed scaling procedure\footnote{Ref.} for an analysis of $M(H)$ curves above and below this phase transition and we find inconsistencies that indicate that the transition is not always related to a melting of the vortex lattice.}

Our scaling procedure (see Refs.\footnote{Ref.}) is based on the fact that, if the Ginzburg-Landau parameter $\kappa$ is temperature independent, the magnetic susceptibility $\chi$ in the mixed state of a type-II superconductor may be written as

$$\chi(H,T) = \chi(H/H_{c2}),$$

where $H_{c2} = H_{c2}(T)$ is the upper critical field. According to Eq. (1), the temperature dependence of $\chi$ is solely determined by the temperature variation of $H_{c2}$. Eq. (1) is sufficient to derive a relation between the magnetizations $M$ at two different temperatures $T$ and $T_0$, which may be written as

$$M(H/h_{c2}, T_0) = M(H, T)/h_{c2}$$

where $h_{c2}(T) = H_{c2}(T)/H_{c2}(T_0)$ is the ratio of the upper critical fields at $T$ and $T_0$. This equation is valid if the diamagnetic response of the mixed state is the only significant contribution to the sample magnetization. It is well known, however, that many superconducting materials, including the HTSC’s that we consider in this work, exhibit sizable paramagnetic susceptibilities in the normal state. In order to account for this additional contribution to the sample magnetization, the following modification of Eq. (2) needs to be made:

$$M(H/h_{c2}, T_0) = M(H, T)/h_{c2} + c_0(T)H.$$  (3)

As is discussed detail in Ref.\footnote{Ref.}, Eq. (3) may be used for the scaling of equilibrium magnetization $M(H)$ curves measured at different temperatures, to obtain $M(H, T)$ curves. The scaling parameters $h_{c2}(T)$ and $c_0(T)$ are determined by the condition that the $M(H, T_0)$ curves, calculated from the magnetization data measured at different temperatures, collapse onto the same master curve. In this way the temperature dependence of the normalized upper critical field $h_{c2}(T)$ and the equilibrium magnetization curve $M_{eff}(H) = M(H, T_0)$ are obtained. While in Refs.\footnote{Ref.}, the main goal was to establish the $h_{c2}(T)$ curves, in the present work we analyze the scaled $M(H)$ curves in the vicinity of the first order phase transition.

In our previous work we discussed in detail the conditions under which our scaling procedure is valid and we showed that these conditions are considerably less restrictive than those that were chosen in previously presented analyses of $M(H, T)$ curves in the mixed state of HTSC’s.\footnote{Our only assumption, the validity of Eq. (1), is rather general and the scaling procedure can be used for the analysis of the equilibrium magnetization data independent of the sample geometry, the pairing type, or of the particular configuration of the mixed state.}

Because the validity Eq. (3) is restricted to equilibrium magnetization, only $M(H, T)$ curves collected above the irreversibility line can be used for the evaluation of the scaling parameters $h_{c2}(T)$ and $c_0(T)$. Nevertheless, some additional information may also be gained from the analysis of scaled $M_{eff}(H)$ curves calculated from magnetization data below the irreversibility line. It was argued in Ref.\footnote{Ref.} that, if the calculated $M_{eff}(H)$ curves collapse onto a single curve also below the irreversibility line, this may be regarded as strong evidence that the corresponding branches of the measured $M(H, T)$ curves...
curves represent the equilibrium magnetization. It was indeed demonstrated in Ref. 3 that for many Bi-based HTSC’s, the $M(H)$ data collected in increasing magnetic fields follow the equilibrium magnetization curve $M_{eq}(H)$ even in fields well below the irreversibility line. Below we show that this is also true for an optimally doped YBa$_2$Cu$_3$O$_{7-x}$ (Y-123) sample, while in La$_2$CuO$_4$ (La-214) based cuprates, this effect is practically unobservable and both branches of the measured $M(H)$ curves deviate from $M_{eq}(H)$ just below the irreversibility line.

We apply the scaling described by Eq. (3) to results of magnetization measurements in the vicinity of the first order phase transition that are available in the literature. It turns out that some of the results of our analysis are in contradiction with the vortex-lattice-melting hypothesis. For this reason, we use the more general nomenclature of "high-field" and "low-field phase", instead of the commonly used notations of vortex liquid and vortex solid.

First, we consider the experimental data for two similar (La$_{0.54}$Sr$_{0.46}$)$_2$CuO$_4$ (La-214) single crystals. The scaling results for these samples, calculated for $T_0 = 14$ K, are shown in Fig. 1. In these two experiments, the magnetization is reversible down to magnetic fields well below the phase transition. The collapse of the data points measured at different temperatures is achieved with the same values of $h_{c2}(T)$ and $c_0(T)$ both below and above the transition, in complete agreement with the expectation outlined above. The results displayed in Fig. 1 may be interpreted as evidence for the existence of two different modifications of the mixed state with two different equilibrium $M_{eff}(H,T_0)$ curves above and below the transition. In the following we use $M_{eq}^{(lp)}$ and $M_{eq}^{(hp)}$ to distinguish the equilibrium $M_{eff}(H)$ curves at $T = T_0$ for the low- and the high-field modifications of the mixed state, respectively. As may be seen in Fig. 1, the low-field modification corresponds to somewhat higher values of the diamagnetic moment (smaller vortex density). The difference $\Delta M = |M_{eq}^{(lp)} - M_{eq}^{(hp)}|$ is positive. Analogous results for optimally doped Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ (Bi-2212) and Y-123 samples are shown in Figs. 2 and 3, respectively. The behavior of the magnetization in the vicinity of the phase transition for these two samples is similar to that presented in Fig. 1. The $M_{eq}^{(lp)}$ and $M_{eq}^{(hp)}$ curves are readily identified, in spite of a pronounced peak effect.

![FIG. 1: The $M_{eff}(H/h_{c2})$ curves for two La-214 samples studied in Refs. 9 and 10. For one of the samples, only reversible magnetization data are shown.](image1)

![FIG. 2: The $M_{eff}(H/h_{c2})$ curves for an optimally doped Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ sample. Only the magnetizations measured in increasing fields are shown. The dashed line indicates the most likely extrapolation of the $M_{eq}^{(lp)}$ curve.](image2)

![FIG. 3: The $M_{eff}(H/h_{c2})$ curves for an optimally doped YBa$_2$Cu$_3$O$_{7-x}$ sample, experimentally investigated in Ref. Only the data collected in increasing fields are shown. The inset shows $M_{eff}(H/h_{c2})$ for $T = 86$ K; the dashed straight lines denote the linear extrapolations of $M_{eq}^{(lp)}(H)$ and $M_{eq}^{(hp)}(H)$.](image3)
observed for the Y-123 sample at the lowest temperature. The difference \( \Delta M = |M_{eq}^{lp}| - |M_{eq}^{hp}| \) is again positive in both cases.

![Graph](image_url)

**FIG. 4:** The \( M_{eff}(H) \) curves for a La-214 sample studied in Ref. [14]. The dashed line indicates \( M_{eq}^{lp}(H) \). The inset shows the original magnetization data. The vertical arrows mark the phase transition, as claimed in Ref. [13].

The \( M_{eff}(H) \) curves in the transition region for another La-214 sample, which are displayed in Fig. 4, are quite different from those shown in Figs. 1-3. The easily distinguishable \( M_{eq}^{lp}(H) \) and \( M_{eq}^{hp}(H) \) curves indicate that the difference \( \Delta M = |M_{eq}^{lp}| - |M_{eq}^{hp}| \) is negative.

The original magnetization data are shown in the inset of Fig. 4: the only difference to Fig. 1 of Ref. [13] is that we use a log-scale for \( H \). The arrows indicate the middle-points of the phase transitions at the corresponding temperatures, as claimed in Ref. [13]. Our plot reveals no evidence for phase transitions at these magnetic fields. We argue that the natural curvature of the \( M(H) \) curves that were plotted on linear scales, emphasized by a real change of slopes at lower fields, lead to a questionable identification of the phase transitions.

Finally we show the result of the scaling procedure for an overdoped Bi\(_2\)Sr\(_2\)CaCu\(_2\)O\(_{8+x}\) sample in Fig. 5. Although only \( M(H) \) data in magnetic fields above the transition were used for the evaluation of \( h_{c2}(T) \) and \( c_0(T) \), the magnetization curves measured in increasing magnetic fields collapse onto a single \( M_{eff}(H) \) curve also below the irreversibility line and thus, as argued above, represent the equilibrium magnetization for \( T = T_0 \). The equilibrium \( M(H) \) curves for the low and high field modifications of the mixed state are such that the difference \( \Delta M = |M_{eq}^{lp}| - |M_{eq}^{hp}| \) in the transition region is again negative. The width of the transition from one modification of the mixed state to the other, which is quite large at higher temperatures, is considerably reduced with decreasing temperature. Also in this case we disagree with the interpretation of the magnetization curves given by the authors of the original publication. According to Ref. [14], the difference \( \Delta M \) changes its sign at \( T \approx 50 \) K, while from Fig. 5 it is clear that \( \Delta M \) is always negative and its absolute value monotonically decreases with decreasing temperature.

The most unexpected result of our analysis is that the magnetization difference \( \Delta M \) across the transition may adopt either sign. This result is difficult to reconcile with the vortex-lattice-melting hypothesis. In case of vortex lattice melting, the external magnetic field acts as pressure does in traditional solid-liquid melting transitions. Thermodynamics requires that the phase corresponding to the higher pressure must have a higher density, independent of whether this high-pressure phase is a liquid or a solid. In relation with the mixed state of type-II superconductors, the vortex liquid necessarily has to adopt a higher vortex density, i.e., the difference \( \Delta M = |M_{eq}^{lp}| - |M_{eq}^{hp}| \) must always be positive. Since negative values of \( \Delta M \) are identified for materials belonging to two different families of HTSC’s, this can hardly be refuted as an accidental result.

In the bulk of the existing literature, the first order transition in the mixed state of HTSC’s is viewed as a melting transition in the system of vortices. In this scenario, the mixed state above the transition represents the vortex liquid, while the vortex solid below the transition is described as a lattice or Bragg glass of vortices, depending on the particular experimental conditions. Numerous experimental observations in the literature are in agreement with this interpretation. However, if the vor-
tex lattice melting is indeed always responsible for the first order transition, all experimental results must find their explanation from this point of view. Apparently, this is not the case. We see no way in which the negative values of $\Delta M$, clearly demonstrated in Figs. 4 and 5, may be explained by invoking the vortex-lattice-melting hypothesis. We also note that in the case of the Y-123 sample, the slope $dM_{eq}/d(\ln H)$ in the low-field phase is substantially smaller than that in the high-field phase (see inset of Fig. 3). Because an order-disorder transition, such as the vortex lattice melting, cannot significantly change the field dependence of the sample magnetization, such a change of $dM_{eq}/d(\ln H)$ is not expected at a vortex lattice-melting-transition.

If there is a first order phase transition in the mixed state of HTSC's which is not related to vortex lattice melting, it must be of different origin. Below we present several possible scenarios. We do not argue in favor of one or the other possibility and we do not even claim that all these scenarios are indeed realistic. Of course, we cannot exclude other possibilities which are not considered here.

1. The change of the symmetry of the vortex lattice. It is well known that with increasing magnetic field a triangular vortex lattice may change to a square one via a first-order phase transition. In general, such a transition reduces the density of vortices. It is also possible, however, that a combination of this transition and a melting of the vortex lattice has to be considered. It is indeed possible that the position of the melting line $H_m(T)$ depends on the symmetry of the vortex lattice and two different melting lines $H_m^{(tr)}(T)$ and $H_m^{(sq)}(T)$ exist for the triangular and square configurations, respectively. If $H_m^{(sq)}(T) < H_{sym}(T) < H_m^{(tr)}(T)$, where $H_{sym}(T)$ denotes the symmetry transition, we have, with increasing field or temperature, a transition to a square vortex lattice which immediately melts down. In this case, there are two contributions to $\Delta M$ with opposite signs and the resulting value of $\Delta M$ may be positive or negative depending on the experimental conditions.

2. A magnetic field induced transition in the superconducting material which is not directly related to the mixed state but changes the superconducting parameters of the sample. It is difficult to imagine, however, that the temperature dependence of such a transition follows that of the first order phase transition in the mixed state of HTSC's.

3. It was suggested in Ref. 13 that in high magnetic fields, the mixed state of high-$\kappa$ superconductors is formed by superconducting filaments in a normal-state matrix, instead of Abrikosov vortices in a superconducting background. The transition to the vortex state with decreasing magnetic field occurs via a topological transition with all generic features of a first order phase transition. The sign of $\Delta M$ would depend on particular experimental conditions.

In conclusion, the most important result of our analysis is that the difference $\Delta M = |M_{eq}^{(lp)}| - |M_{eq}^{(hp)}|$ between the equilibrium magnetization curves below and above the transition in the mixed state of HTSC's may be negative as well as positive. The negative sign of $\Delta M$ is in serious conflict with the interpretation that this transition reflects the melting of the vortex lattice.

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