On the interpretation of the equilibrium magnetization in the mixed state of high-$T_c$ superconductors.

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Abstract

We apply a recently developed scaling procedure to the analysis of equilibrium magnetization $M(H)$ data that were obtained for Tl$_2$Ba$_2$CaCu$_2$O$_{8+x}$ and Bi$_2$Sr$_2$CaCu$_2$O$_8$ single crystals and were reported in the literature. The temperature dependencies of the upper critical field and the magnetic field penetration depth resulting from our analysis are distinctly different from those obtained in the original publications. We argue that theoretical models, which are usually employed for analyses of the equilibrium magnetization in the mixed state of type-II superconductors are not adequate for a quantitative description of high-$T_c$ superconductors. In addition, we demonstrate that the scaled equilibrium magnetization $M(H)$ curve for a Tl-2212 sample reveals a pronounced kink, suggesting a phase transition in the mixed state.

Key words:
high-$T_c$ superconductors, upper critical field, equilibrium magnetization, mixed state
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Measurements of the equilibrium magnetization in the mixed state of type-II superconductors are often used for studying conventional and unconventional superconductivity. This is particularly true for high-$T_c$ (HTSC) superconductors, because of the extremely wide range of magnetic fields in which their magnetization is reversible. The physically meaningful information is not straightforwardly accessible, however, and, in order to estimate critical magnetic fields or characteristic lengths from magnetization measurements, theoretical models have to be invoked. Below we consider several theoretical approaches which are usually employed for the interpretation of corresponding experimental results, and show that the resulting temperature dependencies
of the upper critical field $H_{c2}$ and the magnetic field penetration depth $\lambda$ still leave space for improvement.

The Hao-Clem model [1,2] is most widely used for evaluating essential superconducting parameters, such as the upper critical field $H_{c2}$ and the Ginzburg-Landau parameter $\kappa$ of HTSC’s from magnetization data. Because this model takes into account the spatial variation of the order parameter it is, no doubt, a better approximation to the Abrikosov theory of the mixed state [3] than previously used approaches. Nevertheless, the $\kappa(T)$ curves obtained by employing the Hao-Clem model practically always exhibit a rather strong and unphysical increase of $\kappa$ with increasing temperature [4,5,6,7,8,9,10,11,12,13,14,15,16].

An instructive example of this behavior is provided by Figs. 3(a) and 3(b) in Ref. [4] where equilibrium magnetization data of a Tl$_2$Ba$_2$CaCu$_2$O$_{8+x}$ (Tl-2212) single crystal were presented and analyzed. The quoted figures reveal a rather strong increase of $\kappa$ with increasing temperature and, as a consequence, a very unusual temperature dependence of the upper critical field $H_{c2}$, which is shown in the bottom inset of Fig. 1. As may be seen, $H_{c2}(T)$ resulting for this particular sample is temperature independent below $T \approx 73$ K and exhibits an unphysical divergence at higher temperatures. A very similar behavior of the $H_{c2}(T)$ curve, resulting from the same type of analysis, was also reported for Bi$_2$Sr$_2$CaCu$_2$O$_8$ (Bi-2212) samples [6].

More recent theoretical work [19,20,21] was intended to avoid these inadequacies by taking into account some specific corrections to the sample magnetization that are not accounted for in the Abrikosov theory. However, as we argue below, the situation concerning the temperature dependencies of $H_{c2}$ and $\lambda$ may be improved even further by employing a recently established scaling procedure [22].

Our analysis of original data that were presented and discussed in Refs. [4,6,20] is based on a scaling procedure developed in Ref. [22]. It was shown that if the Ginzburg-Landau parameter $\kappa$ is temperature independent, the equilibrium magnetizations in mixed state of type-II superconductors at two different temperatures are related by

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

with

$$ c_0(T) = \chi_{eff}^{(n)}(T) - \chi_{eff}^{(n)}(T_0). $$

In Eqs. (1) and (2) $h_{c2}(T) = H_{c2}(T)/H_{c2}(T_0)$ represents the normalized upper critical field and $\chi_{eff}^{(n)}(T)$ is the effective magnetic susceptibility of the superconductor in the normal state. $T_0$ is an arbitrary chosen temperature within

\footnote{In the Ginzburg-Landau theory, $\kappa$ is temperature independent, and a slight reduction of $\kappa$ with increasing temperature is predicted by microscopic theories [17,18].}
the covered range of temperatures. The first term on the right-hand side of Eq. (1) is universal for any type-II superconductor, while the second is introduced to account for the temperature dependent paramagnetism of HTSC’s in the normal state. In practice, this second term often also includes a non-negligible contribution arising from the sample holder. In the following we use $M_{\text{eff}}(H) = M(H, T_0)$ to denote the magnetization calculated from measurements at $T \neq T_0$ using Eq. (1). The adjustable parameters $h_{c2}(T)$ and $c_0(T)$ may be established from the condition that the $M_{\text{eff}}$ curves, calculated from measured $M(H)$ data in the reversible regime at different temperatures, collapse onto a single $M_{\text{eff}}(H)$ curve which represents the equilibrium magnetization at $T = T_0$ (see Ref. [22] for details).

There are two adjustable parameters in our scaling procedure whereby the upper critical field $H_{c2}$ represents the natural normalization parameter for all magnetic characteristics of the mixed state and, as stated, $c_0(T)$ is essential to account for any temperature dependence of the paramagnetic susceptibility of HTSC’s in the normal state. An important advantage of our scaling approach is that no particular field dependence of the magnetization has to be assumed a priori and therefore, this procedure may be used for any type-II superconductor, independent of the pairing type, the absolute value of $\kappa$, the anisotropy of superconducting parameters, or the sample geometry. However,
because \( M(H) \) is not postulated, the scaling procedure may only provide the relative temperature variation of \( H_{c2} \) given by the scaling parameter \( h_{c2}(T) \). The success of the scaling procedure described by Eq. (1) in data analyses of a number of different HTSC materials was demonstrated in previous work [22,23,24,25,26]. In addition to the temperature dependence of the normalized upper critical field, also the superconducting critical temperature can be evaluated by extrapolation of the \( h_{c2}(T) \) curve to \( h_{c2} = 0 \). The \( T_c \) values evaluated in this way are always consistent with low field \( M(T) \) curves [22,24].

The normalized upper critical field \( h_{c2}(T) \), obtained via the scaling of \( M(H) \) data for a Tl-2212 single crystal presented in [4], is shown in Fig. 1. As may be seen, \( h_{c2}(T) \) for this sample varies linearly with \( T \) above \( 0.8T_c \approx 82 \text{ K} \). This linearity allows for a quite accurate evaluation of the critical temperature \( T_c \) by extrapolating the \( h_{c2}(T) \) curve to \( h_{c2} = 0 \). The inset of Fig. 1 demonstrates that the value of \( T_c = 103.0 \text{ K} \) resulting from this extrapolation, is well in agreement with the temperature dependence of the low-field magnetization of the same sample. As may be seen in Fig. 1, \( h_{c2}(T/T_c) \) for this Tl-2212 sample is practically identical with the \( h_{c2}(T/T_c) \) curves, presented in Ref. [24], for two underdoped YBa\(_2\)Cu\(_3\)O\(_{7-x}\) (YBCO) samples thus supporting our previous suggestion [24] concerning the universality of the temperature dependence of \( H_{c2}(T) \) for HTSC’s.

The temperature dependence of the scaling parameter \( c_0 \), which is shown in Fig. 2, demonstrates that the paramagnetic contribution to the sample magnetization obeys a Curie-type law in a rather extended temperature range with some deviations at temperatures below 55 K, as well as at temperatures very close to \( T_c \).
The dependence of $M_{\text{eff}}$ on $H/h_{c2}(T)$ that results from our scaling procedure is shown in Fig. 3. Because of the high quality of the experimental data presented in Ref. [4] and the extended covered range of magnetic fields, the scaling is nearly perfect. The $M_{\text{eff}}(H)$ data points, calculated from the measurements at different temperatures, combine to a single curve with virtually no scatter. The remarkable feature of this curve is a pronounced kink at $H/h_{c2} \approx 20$ kOe. This kink clearly indicates a significant change in the properties of the mixed state. Unfortunately, the measurements in Ref. [4] are limited to magnetic fields $H \leq 20$ kOe. Only a limited number of data points, measured at $T \geq 96$ K and $H = 20$ kOe combine to the $M_{\text{eff}}(H)$ curve above the kink. This is why, on the basis of the available data, no definite conclusions concerning this observation can be made. Only additional measurements in higher fields can clarify the situation. If this feature is confirmed by a more detailed study, the kink would definitely reflect some kind of phase transition.

If the Ginzburg-Landau parameter $\kappa$ is temperature independent, as it is assumed in our approach, the magnetic field penetration depth $\lambda(T)$ is inversely proportional to the square root of the upper critical magnetic field, i.e.,

$$\lambda(T)/\lambda(T_0) = \sqrt{H_{c2}(T_0)/H_{c2}(T)}.$$  \hspace{1cm} (3)

The temperature dependence of the normalized penetration depth calculated in this way is shown in Fig. 4. As may be seen in the inset of Fig. 4, for $T \geq 0.75T_c$ the resulting temperature dependence of $\lambda$ is well in agreement with the prediction of the Ginzburg-Landau theory. At lower temperatures, Eq. (3) is not applicable for the evaluation of $\lambda(T)$ and therefore it is not
Fig. 4. The normalized temperature dependence of $\lambda$ calculated from the $h_{c2}(T)$ curve. The two $\lambda(T)$ curves presented in Ref. [4] are shown for comparison. The inset shows the normalized $\lambda$ vs $1/(1-T/T_c)$ on Log-scales; the straight line corresponds to expectations of the Ginzburg-Landau theory.

It is surprising that $\lambda(T)$ deviates from the Ginzburg-Landau type behavior. The plots in Fig. 4 also demonstrate that our $\lambda(T)$ curve is quite different from those calculated in Ref. [4] by employing either a modified London model (nonlocal theory) [21] or the Bulaevskii-Ledvji-Kogan approach (BLK theory) [19]. These differences are particularly pronounced in the temperature range where our $\lambda(T)$ curve matches the Ginzburg-Landau theory.

Next we consider another example to support our arguments. We apply our analysis procedure on experimental $M(H)$ data for a single crystal of Bi$_2$Sr$_2$CaCu$_2$O$_8$, published in Ref. [6]. These results were previously analyzed in Ref. [20] and we compare our results with those presented in [20]. In Fig. 5 we show a comparison of the temperature dependence of the normalized upper critical field obtained from the same experimental data by employing three different approaches.

The plots in Figs. 1, 4 and 5, demonstrate that employing our scaling procedure for the analysis of the equilibrium magnetization data results in conventional temperature dependencies of $H_{c2}$ and $\lambda$ which are rather different from those obtained by invoking other, previously considered approaches [1,2,19,20,21]. The models discussed in [20,21] were specially invented in order to explain the failure of the Hao-Clem model in handling magnetization data for layered HTSC compounds, as exemplified by the bottom inset of Fig. 1, and to improve the interpretation of experimental results. The enhancement of $H_{c2}$ at high temperatures was explained in Ref. [20] by the influence of thermal fluctuations on the sample magnetization, which are not accounted for in...
the Hao-Clem model. The temperature independence of $H_{c2}$ at lower temperatures was interpreted in Ref. [21] in terms of non-local effects. It is argued in [21] that due to effects of non-locality, which are expected to be important at temperatures well below $T_c$, the upper critical field $H_{c2}$ in the expression for the sample magnetization should be replaced by another field $H_0$ which depends on temperature much weaker than $H_{c2}$. The results of our analysis give no support to any of these two assumptions. As was demonstrated above, the sample magnetization can be scaled by simply invoking $H_{c2}(T)$ in a wide temperature range without significant corrections from thermal fluctuations or other sources.

Because the influence of thermal fluctuations on the magnetization of HTSC’s in the mixed state is often overestimated in the literature, we discuss this point in more detail. As may clearly be seen in Fig. 3, if the experimental magnetizations are corrected for the temperature dependence of $\chi_{eff}^{(n)}$, the $M_{eff}(H)$ data points, corresponding to different temperatures, merge onto exactly the same curve. The obvious conclusion is that fluctuation effects do not produce a considerable contribution to the sample magnetization in this rather wide temperatures range. It should be noted, however that, as may be seen in Fig. 2, the $c_0(T)$ data points for the three highest temperatures ($T \geq 97$ K) deviate from the straight line corresponding to the Curie law. It is possible that these deviations are indeed due to thermal fluctuations. \(^2\)

\(^2\) As was pointed out in Ref. [22], at temperatures close to $T_c$, the term $c_0(T)H$ may also account for fluctuation effects. However, because the fluctuation-induced con-
temperatures $T \geq 100$ K $\approx 0.97T_c$, the magnetization data presented in Ref. [4] cannot satisfactorily be scaled using Eq. (1). We consider this failure of the scaling procedure as evidence for the increasing role of fluctuations with increasing temperature. We also note that in Y-123 compounds the impact of fluctuations effects is even weaker and in some cases our scaling procedure could successfully be employed up to temperatures as high as $0.99T_c$ [22].

At present, it is difficult to identify the exact reason for the failure of the Hao-Clem model, an approximation to the Ginzburg-Landau theory of the mixed state, in treating experimental magnetization data. A few possibilities are mentioned below.

1. According to Ref. [29], the Hao-Clem model suffers from general inaccuracies in corresponding calculations. It is also quite possible that the contribution to the sample magnetization due to the normal-state paramagnetism of HTSC’s is not accounted for with sufficient accuracy in the calculations.

2. Not all the assumptions of the model are satisfied in experiments. In particular, the assumed conventional $s$—pairing [1,2], also the basis for numerical calculations of Brandt [30,31], is not compatible with the now accepted $d$—pairing in HTSC’s.

3. More general reasons for the disagreement between the calculated and the experimental magnetization curves cannot be excluded. The argument is based on results of recent magnetization measurements on single crystals of NbSe$_2$ [32], a conventional superconductor without any expectation of unconventional pairing. Because the upper critical field for this superconductor is not very high, the paramagnetic contribution to the sample magnetization may easily be established and accounted for. For magnetic fields $H > 0.6H_{c2}$ (below $0.6H_{c2}$ the $M(H)$ curves are irreversible) the experimental magnetization $M(H)$ curves are linear if plotted versus $\ln H$. Very accurate numerical calculations of Brandt [30,31] for this magnetic field range result in $M(H)$ curves that vary linearly with $H$, however.

In conclusion, by invoking published results of magnetization measurements on Tl-2212 [4] and Bi-2212 [6] samples we demonstrate that our analysis of equilibrium magnetization data results in temperature variations of $H_{c2}$ and $\lambda$ which are rather different from those obtained by employing theoretical approaches that are traditionally used for the interpretation of magnetization measurements in HTSC’s [1,2,19,20,21]. In view of the successful scaling of the magnetization data which is based on a minimum of $a$ $p$riori assumptions and whose validity was convincingly demonstrated in Refs. [22,24,25,32,33], we are confident that the resulting temperature dependencies of the normalized contribution to the sample magnetization is not linear in $H$, it may only approximately be accounted for.
upper critical field presented in Figs. 1 and 5 correctly describe the $H_{c2}(T)$ curves for these HTSC compounds. We also argue that thermal fluctuations have a much weaker impact on the mixed-state magnetization of HTSC’s than is usually believed.

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