Temperature dependence of the upper critical field of high-$T_c$ superconductors from isothermal magnetization data. Application to polycrystalline samples and ceramics.

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Abstract

Using a recently developed scaling procedure that allows to establish the temperature dependence of the normalized upper critical field $h_{c2}(T) = H_{c2}(T)/H_{c2}(T_0)$ from measurements of the equilibrium isothermal magnetization, we analyze experimental data obtained on polycrystalline and ceramic samples. We show that the scaling procedure works quite well in all cases that we have considered. This provides a very strong evidence that the temperature dependencies of $h_{c2}$ are independent of the orientation of an applied magnetic field with respect to the crystallographic axes of these materials.

Key words:
high-$T_c$ superconductors, upper critical field, equilibrium magnetization, mixed state
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1 Introduction

The Ginzburg-Landau (GL) theory predicts that, if the GL parameter $\kappa$ is temperature independent, the magnetic susceptibility $\chi$ of a type-II superconductor in the mixed state is a universal function of $h = H/H_{c2}(T)$ [1], i.e.,

$$\chi(H, T) = \chi(h).$$

(1)
As has recently been shown [2], this offers the possibility to evaluate the temperature dependence of the normalized upper critical field from measurements of the reversible isothermal magnetization in the mixed state without any additional assumptions. According to Ref. [2], the magnetizations of a sample in a fixed field \( H \) at two different temperatures \( T_0 \) and \( T \) are related by

\[
M(H, T_0) = M(h_c^2 H, T) / h_c^2 + c_0(T) H ,
\]

where \( h_{c2} = H_{c2}(T) / H_{c2}(T_0) \). While the first term on the right side of Eq. (2) follows straightforwardly from Eq. (1), the term \( c_0(T) H \) accounts for the sample magnetization arising from any temperature dependent magnetic susceptibility of the superconductor in the normal state. By a suitable choice of the parameters \( h_{c2} \) and \( c_0(T) \), individual \( M(H) \) curves measured at different temperatures may be merged into a single master curve \( M(H, T_0) \). In this way the temperature dependence of the normalized upper critical field \( h_{c2}(T) = H_{c2}(T) / H_{c2}(T_0) \) can be obtained [2].

It is important to point out that the scaling procedure described by Eq. (2) provides values of the upper critical field as it enters the expression for the magnetization in the mixed state. This definition of \( H_{c2} \) is unambiguous and the physical meaning of the upper critical field defined in this way is the same as in the GL theory. In the \( H - T \) phase diagram the \( H_{c2}(T) \) curve represents the upper boundary of the mixed state. This does not necessarily imply that in magnetic fields exceeding \( H_{c2}(T) \), superconductivity in the sample is completely quenched. This applies particularly to high-\( T_c \) superconductors (HTSC’s) where, due to their rather specific properties, superconducting regions may exist in the form of separated islands or layers oriented along the direction of the magnetic field even in magnetic fields \( H > H_{c2}(T) \). This is discussed in more detail in Ref. [2]. In this case, there is no superconducting phase coherence in the direction perpendicular to the magnetic field and therefore, the magnetic flux is distributed between these islands or layers without the formation of a true mixed state.

2 Analysis of experimental data

As in our previous work, we apply our scaling procedure to magnetization measurements available in the literature, but now concentrating on polycrystalline and ceramic samples. The samples characteristics are listed in Table I. Letters in the sample identification denote the chemical element characterizing the corresponding HTSC family. Because the \( h_{c2}(T/T_c) \) curves for oxygen deficient \( \text{YBa}_2\text{Cu}_3\text{O}_{7-x} \) (YBCO-123) and intrinsically underdoped \( \text{Y}_2\text{Ba}_4\text{Cu}_7\text{O}_{15+4x} \) (YBCO-247) samples are quite different from those of all
other HTSC compounds, we present the results of our analysis in two different subsections.

2.1 Y-247 and oxygen deficient Y-123 materials

Fig. 1(a) demonstrates the resulting $M(H)$ curves for $T_0 = 44$ K, as obtained with our scaling procedure for two YBa$_2$Cu$_3$O$_{7-x}$ samples with $x = 0.19$ (Y#2 and Y#3). Although these two samples have the same oxygen content, their relative densities, with 90% for Y#2 and 67% for Y#3, and their average grain sizes are distinctly different [3]. Naturally this leads to noticeably different $M(H)$ curves, as may be seen in Fig. 1(a). In spite of all these differences, the $h_{c2}(T)$ curves for these two samples are practically identical, as is demonstrated in the inset of Fig. 1(a).

![Magnetization data scaled according to Eq (2). (a) Samples Y#2 and Y#3. The inset shows the corresponding $h_{c2}(T)$ data. The solid line is the best linear fit to $h_{c2}(T)$ for $T \geq 45$ K. (b) Sample Y#7.](image)

The scaled $M(H)$ curve for an Y-247 sample (Y#7) is shown in Fig. 1(b). It is obvious that the scaling procedure is not valid at low temperatures and low magnetic fields. This failure is due to the fact that some of the original data points were measured below the irreversibility line and obviously do not correspond to the equilibrium magnetization.

1 Scaling is, however, well obeyed

It is often the case that the equilibrium magnetization below the irreversibility line is taken as the arithmetic mean of the values measured in increasing and decreasing fields, as it follows from the Bean critical state model [4,5]. This model is based on the assumption that the critical current density $j_c$ is independent of the magnetic induction. However, it turns out that the values of $j_c$ for HTSC’s, evaluated by application of the Bean critical-state model, depend on the applied magnetic field quite dramatically [6,7,8,9,10,11,12,13,14]. Yet, for some mysterious reasons it is generally accepted that the Bean model is nevertheless applicable for this class of...
for $HH_c(90.5K)/H_c(T) \geq 10$ kOe. We present this example here in order to show that, if for some reasons the magnetic susceptibility does not follow Eq. (1), it is recognized by our procedure.

![Graph showing temperature variations of $H_c^2$, normalized by $H_c^2(0.9T_c)$, for several oxygen deficient Y-123 and Y-247 samples. The dashed line represents a calculation based on the weak coupling BCS theory [15,16].](image)

Fig. 2. Temperature variations of $H_{c2}$, normalized by $H_{c2}(T = 0.9T_c)$, for several oxygen deficient Y-123 and Y-247 samples. The dashed line represents a calculation based on the weak coupling BCS theory [15,16]

As has been shown in Ref. [2], the $h_{c2}(T)$ curves for the oxygen deficient YBCO-123 samples are linear down to substantially lower temperatures than those for other HTSC’s. This circumstance provides the possibility of a reliable evaluation of $T_c$ by extrapolating the $h_{c2}(T)$ curve to $h_{c2} = 0$ even if the magnetization measurements are limited to temperatures not very close to $T_c$, as may be seen in the inset of Fig. 1(a). The $h_{c2}(T/T_c)$ curves for several underdoped Y-123 samples, together with the results for two Y-247 samples, Y#4 and Y#5, are shown in Fig. 2.

### 2.2 $h_{c2}(T)$ for other HTSC compounds

Fig. 3(a) shows the magnetization data for a Tl-based cuprate, sample Tl#2, scaled according to Eq. (2). The mismatch between the scaled data points originating from $M(H)$ curves measured at different temperatures does not exceed the scatter of the original experimental data, which is obviously higher at higher temperatures. The resulting values of $h_{c2}(T) = H_{c2}(T)/H_{c2}(105K)$ for $T \geq 95$ K are plotted in Fig. 3(b). The data for $T \geq 107$ K, i.e., $T/T_c \geq 0.92$, lie along a straight line, while in a more extended temperature range, $h_{c2}(T)$ may quite well be approximated by a power law of the form

$$h_{c2}(T) = \frac{H_{c2}(T)}{H_{c2}(T_0)} = \frac{1 - (T/T_c)^\mu}{1 - (T_0/T_c)^\mu} \quad (3)$$

superconductors. Fig. 1(b) represents one additional evidence that the Bean model cannot provide a satisfactory quantitative description of the critical state in HTSC’s. 
Fig. 3. (a) Magnetization data for sample Tl#2 scaled according to Eq. (2) with $T_0 = 105$ K. (b) $H_{c2}(T)/H_{c2}(105K)$ for the same sample. The solid line is the best fit to $h_{c2}(T)$ with Eq. (3) for $T \geq 95$ K. The dashed line is the best linear fit to $h_{c2}(T)$ for $T \geq 107$ K. The inset shows the temperature derivative of the low-field magnetization and the zero-field resistance curves. The vertical line in the inset indicates the position of $T_c$ evaluated by the extrapolation of the $h_{c2}(T)$ curve.

with two fit parameters $T_c$ and $\mu$. Because the magnetization was measured up to temperatures close to $T_c$, both the linear and the power-law extrapolation lead to practically the same value of $T_c$. In cases where the values of $h_{c2}$ sufficiently close to $T_c$ are not available, we employ Eq. (3) for the extrapolation of the $h_{c2}(T)$ curves to $h_{c2} = 0$.

In the inset of Fig. 3(b) the vertical solid line indicates the position of $T_c$ evaluated with our scaling procedure with respect to experimental $dM/dT$ and $R(T)$ curves for the same sample. Note that $T_c$ does not coincide with any distinct point of these curves, such as the temperature where $R$ reaches zero or the maximum of the $dM/dT$ versus $T$ curve.

Fig. 4. (a) and (b) Temperature variations of $H_{c2}$ normalized by $H_{c2}(T = 0.9T_c)$ for different samples listed in Table I. The solid lines represent an analogous curve drawn in Fig. 3(b) of Ref. [2]. The dashed line represents a calculation that is based to the weak coupling BCS theory [15,16].
For the procedure outlined above, as well as in our previous work [2], we have chosen only data from magnetization measurements that were made up to temperatures \( T \sim 0.94 - 0.95T_c \), in order to obtain reliable values of \( T_c \) by extrapolating the \( h_{c2}(T) \) curves. A considerable number of studies available in the literature does not satisfy this criterion, however. As an example we show, in Fig. 5(a), the \( h_{c2}(T) \) curve for a ceramic sample of \( \text{Bi}_1\text{.95Sr}_{2.05-x}\text{La}_x\text{CuO}_y \) (Bi#3). This curve is qualitatively very similar to those shown in Fig. 4 and in a next step we checked whether these and other analogous data may quantitatively be fit by the same solid lines as drawn in Figs. 4(a) and 4(b), simply by adjusting the value of \( T_c \). The result of this procedure for several samples is shown in Fig. 5(b). Again all the \( h_{c2}(T/T_c) \) data follow the same solid line quite closely. The arrow in the inset of Fig. 6 indicates the position of \( T_c \) determined in this way with respect to the field-cooled magnetization curve of sample Bi#3. We applied this kind of analysis also to a number of magnetization studies of single crystals and grain-aligned samples [26,27,28,29,30,31,32,33,34,35,36,37,38,39,40,41]. Again in all these cases, the \( h_{c2}(T) \) curves may very well be fitted to the universal \( h_{c2}(T/T_c) \) curve shown in Fig. 4(b) and originally displayed in Fig. 3(b) of Ref. [2].
3 Conclusion

It turns out that the scaling procedure described by Eq. (2) works quite well not only for single crystals or grained-aligned samples of HTSC’s, as previously demonstrated in Ref. [2], but also for HTSC polycrystals and ceramics. Because polycrystalline samples can be made much larger than single crystals, the magnetization measurements can, in principle, be made more accurately and may easily be extended up to temperatures very close to $T_c$. The fact that our scaling procedure works for polycrystalline or ceramic samples means that the shape of the normalized $H_{c2}(T)$ curves does not depend on the orientation of the external magnetic field with respect to the crystallographic axes. This is undoubtedly the most important result of this study.

The results of our analysis presented in this work confirm the main result of our previous study where it was shown that the normalized temperature variations of the upper critical field follow the same universal curve for most of the HTSC’s [2]. It now appears that all HTSC’s may be divided into two groups with two distinctly different normalized $H_{c2}(T)$ curves (see Figs. 2 and 4). For the smaller group consisting of Y-247 and oxygen deficient Y-123 compounds the temperature dependence of $H_{c2}$ is quite close to that calculated on the basis of the weak-coupling BCS theory [15,16]. For the other group, which includes practically all other HTSC compounds, all the normalized $h_{c2}(T/T_c)$ curves are identical but are well below the calculated values of Refs. [15,16]. At present, it is not clear what causes the difference between the two sets of $h_{c2}(T/T_c)$ curves. We may only state that $h_{c2}(T/T_c)$ of a $\text{YBa}_2\text{Cu}_3\text{O}_{6.85}$ sample is already completely different from that of optimally doped Y-123 material.
Table 1
Sample identification.

| No. | Refs. | Compound                  | $T_c$ (K) |
|-----|-------|---------------------------|-----------|
| Y# 1| [3]   | YBa$_2$Cu$_3$O$_{6.98}$   | 92.1      |
| Y# 2| [3]   | YBa$_2$Cu$_3$O$_{6.81}$   | 62.0      |
| Y# 3| [3]   | YBa$_2$Cu$_3$O$_{6.81}$   | 62.0      |
| Y# 4| [3]   | YBa$_2$Cu$_3$O$_{6.69}$   | 55.9      |
| Y# 5| [17]  | YBa$_2$Cu$_3$O$_{6.5}$    | 44.8      |
| Y# 6| [13]  | Y$_2$Ba$_4$Cu$_7$O$_{15+x}$ | 93.1      |
| Y# 7| [18]  | Y$_2$Ba$_4$Cu$_7$O$_{15+x}$ | 91.6      |
| Y# 8| [18]  | YBa$_2$Cu$_4$O$_8$        | 80.7      |
| Y# 9| [18]  | YBa$_2$Cu$_3$O$_{7−x}$    | 92.9      |
| Y#10| [19]  | YBa$_2$Cu$_3$O$_{7−x}$    | 92.1      |
| Y#11| [20]  | YBa$_2$Cu$_3$O$_{7−x}$    | 92.1      |
| Bi# 1| [21] | Bi$_{2.12}$Sr$_{1.9}$Ca$_{1.2}$Cu$_{1.96}$O$_{8+x}$ | 86.1      |
| Bi# 2| [18] | Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ | 93.2      |
| Bi# 3| [22] | Bi$_{1.95}$Sr$_{2.05−x}$La$_x$CuO$_y$ | 36.8      |
| Tl# 1| [23] | Tl$_2$Ba$_2$Ca$_2$Cu$_3$O$_{10+x}$ | 122.8     |
| Tl# 2| [24] | Tl$_{0.7}$Pb$_{0.2}$Bi$_{0.2}$Sr$_{1.8}$Ba$_{0.2}$Ca$_{1.9}$Cu$_{3}$O$_{10+x}$ | 122.8     |
| Tl# 3| [18] | Tl$_2$Ba$_2$CaCu$_2$O$_{8+x}$ | 107.1     |
| Tl# 4| [18] | Tl$_2$Ba$_2$Cu$_2$O$_{6+x}$ | 92.1      |
| Tl# 5| [18] | Tl$_2$Ba$_2$Cu$_2$O$_x$    | 90.0      |
| Tl# 6| [6]  | Tl$_{0.5}$Pb$_{0.5}$Sr$_4$Cu$_2$CO$_3$O$_7$ | 69        |
| Hg# 1| [6]  | Hg$_2$Ba$_2$Ca$_2$Cu$_3$O$_x$ | 133.0     |

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