Inferring equilibrium magnetization from hysteretic *M-H* curves of type-II superconductors

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Isothermal M-H curves, coupled with the critical state model, are routinely used to extract critical current density $J_c(B)$; and the limitations and validity are well understood. These hysteretic M-H curves can also be used to estimate the equilibrium magnetization $M_{\rm eq}(H)$, and this paper discusses the validity of such a procedure using analytically tractable models for $J_c(H)$. We put special emphasis on the case where the M-H curve shows a fish tail or peak effect, and an experimental procedure to estimate errors in the inferred $M_{\rm eq}(H)$ is presented. The need to infer $M_{\rm eq}(H)$ is underscored by recent experimental works speculating on thermodynamic phase transitions between vortex phases having intrinsic pinning. [S0163-1829(99)01413-7]

Hysteresis is observed in the isothermal M-H curves of most superconductors due to the pinning of vortices. This hysteresis was first related to the critical current density J_c by Bean's critical state model[1] (CSM). The original work assumed a lower critical field $H_{c_1}=0$ and thus ignored the equilibrium magnetization $M_{\rm eq}(H)$. Bean considered an infinitely long cylinder of transverse dimension 2D in a parallel field and assumed field-independent J_c. The field profiles $B(x)$ are then straight lines, and the envelope hysteresis curves [which correspond to the field change having fully penetrated the sample such that $B(x)$ varies monotonically from the surface to the center] are lines of constant *M*, symmetric about $M=0$, with magnitude $M_s=(k/2)J_cD$. Here k is a constant that depends on the shape of the cylinder's cross section. When the actual $M_{\rm eq}(H)$ are included, the field profiles $B(x)$ retain their shape but are shifted to have a value $\mu_0[H+M_{\rm eq}(H)]$ at the surface.[2] Denoting the magnetization in increasing and decreasing field by $M\uparrow(H)$ and $M\downarrow(H)$, we have $M\uparrow(H)=M_{\rm eq}(H)-(k/2)J_cD$ and $M\downarrow(H)=M_{\rm eq}(H)+(k/2)J_cD$, and the hysteresis curves are symmetric about $M_{\rm eq}(H)$. It follows that

$$M_{\rm eq}(H)=\frac{1}{2}[M\uparrow(H)+M\downarrow(H)] \tag{1}$$

and

$$J_c(H)=\frac{1}{kD}[M\downarrow(H)-M\uparrow(H)]. \tag{2}$$

Equation (2) has been assumed to be valid even when J_c depends on the local field *B*, and has been used extensively to infer $J_c(B)$ from the magnetization hysteresis $\Delta M(H)=M\downarrow(H)-M\uparrow(H)$ at $H=B/\mu_0$.

The validity of Eq. (2) for a field dependent $J_c(B)$ was examined by Fietz and Webb.[3] Using a Taylor series expansion, they showed that the correction terms are of order (d^2J_c/dB^2) and higher. Its usage in the high-T_c superconductors surprisingly resulted in field independent J_c at low fields. This was attributed[4] to the breakdown of Taylor series expansion for fields below the field for first full penetration H_I. The applicability of Eq. (2) has in recent years been studied in great detail[4–7] for $J_c(B)$ that decreases with increasing $|B|$. Studies have recently been initiated[8] for situations where J_c vs B shows a ''peak effect.''

The applicability of Eq. (1) has, to our knowledge, not been examined in great detail. Equation (1) would be exact only if the M-H curve is symmetric about $M_{\rm eq}(H)$, and this is not valid if J_C is a function of *B*. Following the Taylor series expansion of Ref. 3, one sees that correction terms will be of order (dJ_c/dB) and errors in inferring $M_{\rm eq}(H)$ will be larger than in inferring J_c. The need for extracting $M_{\rm eq}(H)$ from hysteretic M-H curves is seen for high-T_c as well as some low T_c superconductors which show intrinsic pinning. In materials like Bi-Sr-Ca-Ca-O, Nd-Ce-Cu-O, Y-Ba-Cu-O, and $CeRu_2$,[9–12] there now exist speculations of thermodynamic phase transitions involving phases with intrinsic pinning. Such phase transitions are expected to have characteristic signatures in $M_{\rm eq}(H)$.

In this paper we shall present general intuitive arguments to obtain upper bounds $\Delta(H)$ on the errors in the use of Eq. (1). We shall then consider an analytically tractable model for $J_c(B)$ exhibiting a peak effect. The actual error in the use of Eq. (1) will be obtained for model parameters, and compared with the upper bounds. An experimental method for obtaining these upper bounds will then be presented.

Generalizations of CSM for $J_c(B)$ decreasing monotonically with increasing *B* exist for many functional forms of $J_c(B)$, the most common being the Kim-Anderson and the exponential models.[4–6] Analytical solutions, assuming $H_{c_1}=0$, exist for infinite cylinders in parallel field geometry which have a demagnetization factor $N=0$. While field profiles $B(x)$ do not depend on the shape of the cylinder's cross-section, the magnetization values do.[7] Results are usually presented for the case of an infinite slab in parallel field as this geometry has the simplest algebra. Calculations for other shapes are tedious but straightforward, and since no special features appear in the M-H curves, we shall in this paper present results only for the slab geometry. If we use the M-H curves so obtained, along with Eq. (1), to estimate $M_{\rm eq}(H)$, we will make an error $\delta M_{\rm eq}(H)=1/2[M\uparrow(H)+M\downarrow(H)]-M_{\rm eq}(H)$. In our calculation we shall continue with the assumption $H_{c1}=0$ followed in most papers on the CSM, thus implying $M_{\rm eq}(H)=0$. We will then estimate the error in the use of Eq. (1) from our model calculations, as

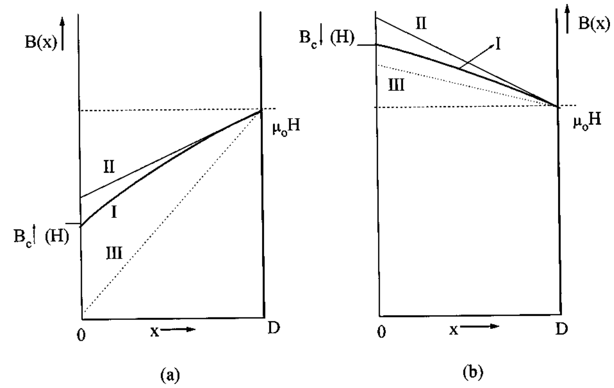

FIG. 1. (a) A schematic plot of the field distribution used in obtaining $M\uparrow(H)$, $M^{\mathrm{MB}}\uparrow(\mu_0 H)$, and $M^{\mathrm{MB}}\uparrow(B_c(H))$ is shown by a thick line (I), a thin line (II), and a dotted line (III), respectively, when the applied field $\mu_0 H$ is increasing. (b) The field distribution case when the applied field $\mu_0 H$ is decreasing used in obtaining the magnetization $M\downarrow(H)$, $M^{\mathrm{MB}}\downarrow(\mu_0 H)$ and $M^{\mathrm{MB}}\downarrow(B_c(H))$ is shown by a thick line (I), a thin line (II), and a dotted line (III), respectively.

$$\delta M_{\mathrm{eq}}(H)=\frac{1}{2}[M\uparrow(H)+M\downarrow(H)]. \qquad (3)$$

We shall show in the Appendix that our error estimates remain accurate for nonzero $M_{\mathrm{eq}}(H)$ in the limit $H \gg H_{C1}$.

We now address the question of estimating $\delta M_{\mathrm{eq}}(H)$ without knowing the detailed form of $J_c(B)$. In Figs. 1(a) and 1(b) we show the field profiles, at $H>H_I$, for the field increasing and decreasing case, respectively. The slope of the profile varies from point to point and equals the J_c at that B. The simplicity of algebra in the slab geometry results in the magnetization being simply proportional to the area contained between the field profile $B(x)$ and the horizontal line $B=\mu_0 H$ in each case. Because of the field dependence of $J_c(B)$, the areas corresponding to $M\uparrow(H)$ and $M\downarrow(H)$ are not equal in magnitude, and the error $\delta M_{\mathrm{eq}}(H)$ is thus nonzero. The question we address is whether this difference can be related to measurable quantities. We denote by $B_c\uparrow(H)$ the field at the center of the sample when the applied field H is increasing, and note from Fig. 1(a) that $J_c(B_c\uparrow(H))$ is the largest slope $B(x)$ has. In what is sometimes referred to as the modified Bean model[13] (MB), we can calculate magnetization $M(H)$ assuming that $B(x)$ are straight lines with slope dictated by $J_c(\mu_0 H)$. Referring to this approximation as $M^{\mathrm{MB}}(H)$, we note that $M^{\mathrm{MB}}\uparrow(H) = -M^{\mathrm{MB}}\downarrow(H) = -(k/2)J_c(\mu_0 H)D$ and $k=1$ for a slab geometry. This is shown schematically in Fig. 1(a) where the thin line (marked II) gives $B(x)$ if we assume $J=J_c(\mu_0 H)$ everywhere. The area enclosed between this thin line and $B=\mu_0 H$ (dashed horizontal line) gives the magnetization $M^{\mathrm{MB}}\uparrow(H)$. In the same figure, the dotted line (marked III) shows $B(x)$ if we assume that $J=J_c(B_c\uparrow(H))$ everywhere, and the area enclosed between this line and $B=\mu_0 H$ gives the magnetization $M^{\mathrm{MB}}\uparrow[B_c\uparrow(H)]$. It is then easy to see that $M^{\mathrm{MB}}\uparrow(B_c\uparrow(H))<M\uparrow(H)<M^{\mathrm{MB}}\uparrow(H)$. Using similar arguments and Fig. 1(b), we note that $M^{\mathrm{MB}}\downarrow(B_c\downarrow(H)) < M\downarrow(H)<M^{\mathrm{MB}}\downarrow(H)$. Combining these inequalities, we get $0>\frac{1}{2}[M\uparrow(H)+M\downarrow(H)]>(k/4)D[J_c(B_c\downarrow(H)) - J_c(B_c\uparrow(H))]$. On using Eqs. (2) and (3), and defining $\Delta(H)=\frac{1}{4}[\Delta M(B_c\downarrow(H))-\Delta M(B_c\uparrow(H))]$, we get,

$$|\delta M_{\mathrm{eq}}(H)|<|\Delta(H)|. \qquad (4)$$

Inequality (4) thus puts an upper bound on the errors in terms of the $\Delta M(H)$ measured in the *same* experiment. We shall describe later how $B_c\uparrow(H)$ and $B_c\downarrow(H)$ can be experimentally estimated.

We now propose an analytically tractable model for a peak effect in $J_c(B)$ as

$$J_c(B)=\begin{cases} J_c(0)\exp(-B/\mu_0 H_0) & \text{for } 0<B<B_1, \\ J_c(0)\exp\left(\dfrac{B-B_1}{\mu_0 H_1}-\dfrac{B_1}{\mu_0 H_0}\right) & \text{for } B_1<B<B_2, \\ J_c(0)\exp\left(\dfrac{2B_2-B_1}{\mu_0 H_1}-\dfrac{B_1}{\mu_0 H_0}-\dfrac{B}{\mu_0 H_1}\right) & \text{for } B>B_2. \end{cases} \qquad (5)$$

Here $J_c(B)$ shows a peak at B_2 around which it falls symmetrically with a decay constant $\mu_0 H_1$. The peak is initiated at B_1. The limit of large B_1 gives us a monotonic exponentially decaying $J_c(B)$. To calculate M-H curves for this model, we follow the methods described earlier.[7,15] We first define a generalized field variable[14] (with dimension of length) $h(B)=\int_0^B dB/(\mu_0 J_c(B))$. The magnetization is then obtained as[7,15] $M\uparrow(H)=-H+\int_{h(B_c\uparrow(H))}^{h(\mu_0 H)}B(h)dh/(\mu_0 D)$ and $M\downarrow(H)=-H+\int_{h(\mu_0 H)}^{h(B_c\downarrow(H))}B(h)dh/(\mu_0 D)$ where $B(h)$ will be obtained by inverting $h(B)$. The advantage of using the variable h is that[7,15] $h(B_c\uparrow(H))=h(\mu_0 H)-D$; and $h(B_c\downarrow(H))=h(\mu_0 H)+D$. If we now define $G(h)=\int_0^h B(h)dh$, we get

$$M\uparrow(H)=-H+\frac{1}{\mu_0 D}[G(h(\mu_0 H))-G(h(\mu_0 H)-D)],$$

$$M\downarrow(H)=-H+\frac{1}{\mu_0 D}[G(h(\mu_0 H)+D)-G(h(\mu_0 H))], \qquad (6)$$

and we also get $\Delta M(H)$ analytically.

For the model defined by Eq. (5), $h(B)$, $B(h)$, and $G(h)$ are all obtained trivially. The results for $G(h)$ are given below

$$G(h)=\mu_0 H_0\left[\left(h+\frac{H_0}{J_c(0)}\right)\ln\left(\frac{J_c(0)}{H_0}h+1\right)-h\right]$$

$$\text{for } 0<h<h(B_1), \quad (7a)$$

$$\begin{aligned}G(h)=&G(h(B_1))+(B_1+\mu_0H_1)(h-h(B_1))-\mu_0H_1\\&\times\left[h-\frac{H_1}{J_c(0)}\exp\left(\frac{B_1}{\mu_0H_0}\right)-h(B_1)\right]\\&\times\ln\left(1-\frac{(h-h(B_1))J_c(0)}{H_1\exp(B_1/\mu_0H_0)}\right)\end{aligned}$$

$$\text{for } h(B_1)<h<h(B_2), \quad (7b)$$

and for $h>h(B_2)$,

$$\begin{aligned}G(h)=&G(h(B_2))-\mu_0H_1(h-h(B_2))\\&-\frac{H_1B_2}{J_c(0)}\exp\left(\frac{B_2}{\mu_0H_1}\right)\exp\left(\frac{B_1}{\mu_0H_0}-\frac{2B_2-B_1}{\mu_0H_1}\right)\\&+\mu_0H_1\left[(h-h(B_2))+\frac{H_1}{J_c(0)}\right.\\&\left.\times\exp\left(\frac{B_2}{\mu_0H_1}\right)\exp\left(\frac{B_1}{\mu_0H_0}-\frac{2B_2-B_1}{\mu_0H_1}\right)\right]\\&\times\ln\left[\frac{J_c(0)}{H_1}(h-h(B_2))\right.\\&\left.\times\exp\left(\frac{2B_2-B_1}{\mu_0H_1}-\frac{B_1}{\mu_0H_0}\right)+\exp\left(\frac{B_2}{\mu_0H_1}\right)\right]. \quad (7c)\end{aligned}$$

The M-H curves given by Eqs. (6) and (7) are thus obtained analytically. One example is plotted in Fig. 2 for the parameters $\mu_0H_0=\mu_0H_1=0.2$, $B_1=0.6$, $B_2=0.8$, and $\mu_0J_c(0)D=0.1$ (magnetization M and the fields B and H are in MKS units). We use Eq. (3) and also plot the errors δM_{eq} is Fig. 2. And we also plot in Fig. 2 the upper bounds $\Delta(H)=[\Delta M(B_c\downarrow(H))-\Delta M(B_c\uparrow(H))]/4$. We have confirmed from our results for various values of the parameters that inequality (4), viz. $|\delta M_{\mathrm{eq}}(H)|<|\Delta(H)|$ is satisfied for both monotonic exponential $J_c(B)$, and for $J_c(B)$ showing differing extents of the peak effect. Before initiating a discussion on the experimental method of obtaining $B_c\uparrow(H)$ and $B_c\downarrow(H)$, we wish to point out that inequality (4) can be violated only when there is a gross violation of Eq. (2). As noted earlier, this can happen only when a Taylor series expansion for $B(x)$ breaks down[4] and that is when $B(x)$ has an inflexion point. Since $J_c(B)$ is small at B_1, this can occur only in a very narrow range of fields near B_1. Our results however show no evidence of inequality (4) breaking down near B_1. It is to be noted from Fig. 2 that $\delta M_{\mathrm{eq}}(H)$ is of the order of a few percent of the hysteresis $\Delta M(H)$, and the upper bound $\Delta(H)$ overestimates $\delta M_{\mathrm{eq}}(H)$ by up to a factor of 2.

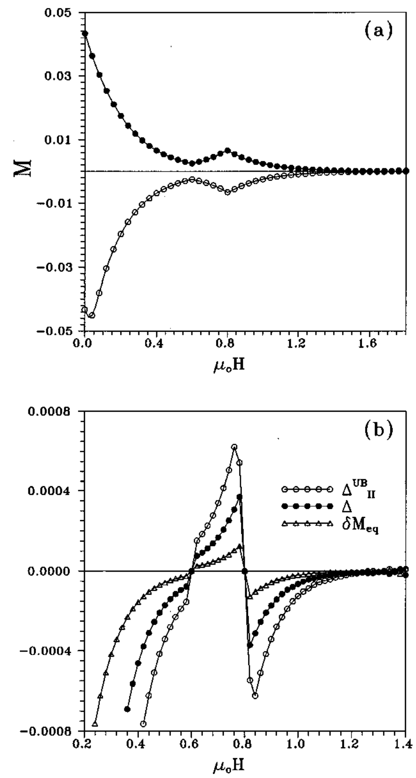

FIG. 2. (a) The envelope M-H curves with nonmonotonically varying $J_c(B)$ with $B_1=0.6$, corresponding to $J_c(B_2)/J_c(0)=e^{-2}$. Since the CSM has the symmetry $M\uparrow(-H)=-M\downarrow(H)$, we shall show M-H curves only for positive H. (b) We plot $\delta M_{\mathrm{eq}}(H)$ and $\Delta(H)$. We also show $\Delta_{II}^{UB}(H)$ which is measurable isothermally. See text for details.

Once isothermal M-H curves are measured, $M_{\mathrm{eq}}(H)$ can be estimated from Eq. (2) and for error bars $\delta M_{\mathrm{eq}}(H)$ we require to use Eq. (4). The only information not already contained in the M-H curves is a knowledge of $B_c\uparrow(H)$ and $B_c\downarrow(H)$. For any field H these can be estimated as follows. After measuring the M-H envelope curves at any temperature T_0, field cool the sample from above T_c to T_0 in field H. Then isothermally reduce the field while measuring the magnetization. It will merge with the envelope $M\downarrow(H)$ curve at $B_c\uparrow(H)$.[15,16] Similarly, after field cooling the sample to T_0 in field H, one should measure the magnetization while raising the field. It will merge with the envelope $M\uparrow(H)$ curve at $B_c\downarrow(H)$. Since $B_c\uparrow(H)$ and $B_c\downarrow(H)$ are now known, the upper bound $\Delta(H)$ can be known from the M-H curves.

Field-cooled measurements are usually more tedious than isothermal measurements. In an isothermal measurement if one starts from the field-increasing envelope curve $M\uparrow(H)$ and starts reducing the field, the minor loop will merge with the field-decreasing envelope curve $M\downarrow$ at $B_{II}\uparrow(H)$, where $B_{II}\uparrow(H)<B_c\uparrow(H)$.[15] Similarly, by starting from $M\downarrow(H)$ and raising the field, the minor loop will merge with the field-increasing envelope curve at $B_{II}\downarrow(H)$, where $B_{II}\downarrow(H)>B_c\downarrow(H)$. And as long as $\Delta M(H)$ is monotonic between $B_{II}\uparrow(H)$ and $B_{II}\downarrow(H)$, we can replace $B_c\uparrow(H)$ by $B_{II}\uparrow(H)$ and $B_c\downarrow(H)$ by $B_{II}\downarrow(H)$ in inequality given by Eq. (4). We note that,[15] $h(B_{II}\uparrow(H))=h(\mu_0H)-2D$, and $h(B_{II}\downarrow(H))=h(\mu_0H)+2D$. We denote the upper bound obtained using these fields by $\Delta_{II}^{UB}(H)$. Since these isothermal measurements are more convenient, we have also compared in Fig. 2 our calculated $\Delta_{II}^{UB}(H)$ where $B_c\uparrow$ and $B_c\downarrow$ are replaced by $B_{II}\uparrow$ and $B_{II}\downarrow$, respectively. As expected, $\Delta_{II}^{UB}(H)$ is larger in magnitude than $\Delta(H)$ over most of the field region. We thus have a completely isothermal technique of estimating $M_{\mathrm{eq}}(H)$ along with error bars $\delta M_{\mathrm{eq}}(H)$. This technique has been used in a recent experiment to estimate $M_{\mathrm{eq}}(H)$ in $CeRu_2$,[12] and the upper bound $\Delta_{II}^{UB}(H)$ on $\delta M_{\mathrm{eq}}(H)$ were negligible compared to $M_{\mathrm{eq}}(H)$.

To conclude, we have in this paper investigated in detail the errors in estimating $M_{\mathrm{eq}}(H)$ from isothermal M-H curves. We have solved analytically a model for the case where a fishtail or peak effect is seen. In view of recent speculations[9–12] of thermodynamic phase transitions at the onset of the fishtail or the peak effect, equilibrium magnetization is a very important thermodynamic parameter. Our analysis has concluded with an experimental technique of providing an upper bound on the errors in estimating $M_{\mathrm{eq}}(H)$.

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APPENDIX

We take $H_{C1}\neq 0$ and $M_{\mathrm{eq}}(H)\neq 0$ and following pages 85–88 of de Gennes[2] set $B=B_{\mathrm{eq}}(H)=\mu_0(H+M_{\mathrm{eq}}(H))$ at the surface of the slab. We denote the magnetization then obtained by $m(H)$, and the magnetization obtained with the assumption $H_{C1}=0$ by $M(H)$. A field-dependent $J_C(B)$ is assumed. A look at Figs. 3.13(b), 3.14, and 3.16 of Ref. 2 immediately gives us [note that $M_{\mathrm{eq}}(H)$ is negative], $m\uparrow(H)=M_{\mathrm{eq}}(H)+M\uparrow(h)$, and $m\downarrow(H)=M_{\mathrm{eq}}(H)+M\downarrow(h)$, where $h=H+M_{\mathrm{eq}}(H)$. We then get, $1/2[m\uparrow(H)+m\downarrow(H)]-M_{\mathrm{eq}}(H)=1/2[M\uparrow(h)+M\downarrow(h)]$, or $\delta m_{\mathrm{eq}}(H)=\delta M_{\mathrm{eq}}(h)$, where $\delta M_{\mathrm{eq}}(h)$ is the asymmetry about $M=0$ when we assume $H_{C1}=0$, and $\delta m_{\mathrm{eq}}(H)$ is the asymmetry about $M_{\mathrm{eq}}(H)$ in a ‘‘proper’’ calculation.

By assuming $H_{C1}=0$, and thereby ignoring the difference between the applied field and the surface field, we only displaced the asymmetry at H to $h=H+M_{\mathrm{eq}}(H)$. The effect is negligible as long as $M_{\mathrm{eq}}\ll H$, which is much weaker than $H_{C1}\ll H$.

We note that we have, following standard treatments of the CSM, ignored surface barrier effects. These are important only at low fields.[17]

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