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Reversible magnetization of high-T_c materials in intermediate fields

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In high-T_c superconductors there exists a broad field domain, in which the reversible magnetization **M** is linear in the logarithm of the applied field H_a. The dependence $\mathbf{M}(\ln H_a)$ is obtained taking the strong uniaxial anisotropy into account. It is shown, that for an arbitrary orientation of the single crystal with respect to $\mathbf{H}_a$ the magnetization vector has a component normal to the applied field comparable to the usually measured component parallel to $\mathbf{H}_a$. A procedure is suggested for extracting the value of the penetration depth from the linear part of $\mathbf{M}(\ln H_a)$. Being applied to the data available for $Y_1Ba_2Cu_3O_7$, the method yields $\lambda \propto (T_c - T)^{-1/2}$ near T_c up to temperatures $(T_c - T)/T_c \sim 10^{-3}$. This puts an upper bound upon the value of the temperature domain, in which the underlying mean-field theory is valid.

The penetration depth λ of all known high-T_c superconductors exceeds by a factor of 100 their coherence length ξ. There exists in these materials a broad field domain

$$H_{c1} \ll H \ll H_{c2}\,, \tag{1}$$

in which the average intervortex distance L obeys the inequality $\xi \ll L \ll \lambda$. The free-energy density F in this domain can be evaluated using the London approach as is done in Ref. 1:

$$8\pi F = B^2 + (B\phi_0/2\pi\lambda^2)\ln(L\beta/\xi)\,. \tag{2}$$

Here B is the magnetic induction, ϕ_0 is the flux quantum, and β is a constant of the order unity, which depends upon the flux line lattice structure (and upon a particular cutoff chosen to evaluate a logarithmically divergent sum over the reciprocal-lattice space). The average spacing between flux lines is defined by $L^2 = \phi_0/B$. The logarithm in Eq. (2) can be written as $\frac{1}{2}\ln(\beta_1 H_{c2}/B)$ with another constant β_1. Then, the field H is readily found:

$$H = 4\pi\partial F/\partial B = B + (\phi_0/8\pi\lambda^2)\ln(\beta_2 H_{c2}/B)$$

with $\beta_2 = \beta_1/e$. The pre-ln factor is of the order H_{c1}; i.e., it is small as compared to both H and B. One can, therefore, replace B with H under the ln sign and obtain for the magnetization M:

$$-4\pi M = (\phi_0/8\pi\lambda^2)\ln(H_{c2}\beta/H)\,. \tag{3}$$

The subscript is omitted in the unknown parameter β; we consider β in the following as a parameter to be found from the experimental data.

Equation (3) represents the constitutive relation for a pinning free isotropic superconductor in the field domain (1). The actual region of its validity might be more narrow. If $H' > H_{c1}$ is a minimum field, at which effects of irreversibility can be ignored, the field domain, where Eq. (3) holds, is given by

$$H' < H \ll H_{c2}\,. \tag{4}$$

It is worth noting that in intermediate fields (1) the demagnetization effects are weak. For an ellipsoidal sample, e.g., the relation between the applied field $\mathbf{H}_a$ and the internal field $\mathbf{H}$ reads $H_{ai} = H_i + 4\pi N_{ik}M_k$, where N_{ik} is the demagnetization tensor.[2] Because $M \sim H_{c1}$, the difference between H_a and H is small; therefore, the field H in Eq. (3) can be considered as the applied field. Thus, in the domain (1) the *reversible* magnetization should be linear in the logarithm of the applied field.

The result (3) for $M(H)$ holds for an isotropic material, while all known high-T_c superconductors are strongly anisotropic. Although most of them are orthorhombic, the anisotropy between the $\hat{\mathbf{c}}$ crystal direction (the long side of the primitive cell) and either $\hat{\mathbf{a}}$ or $\hat{\mathbf{b}}$ (in the Cu-O plane) is much larger than a relatively small "in-plane" anisotropy. Even if the in-plane anisotropy is not weak, it would be masked by the presence of twin domains with interchanging $\hat{\mathbf{a}}$ and $\hat{\mathbf{b}}$ directions. Therefore, to describe major anisotropy effects, one can consider these materials as being uniaxial.

The anisotropic generalization of the London equations can be obtained by replacement of an isotropic mass M ($\lambda^2 \propto M$) with a mass tensor M_{ik}.[3,4] (The notation "M" for the mass is used only in this paragraph; it should not be confused with that of the magnetization.) Following the formal procedure of Ref. 1 of the isotropic case, one obtains for the free energy of a lattice of vortices inclined at an angle θ with respect to the $\hat{\mathbf{c}}$ axis of the crystal:[5]

$$8\pi F = B^2 + (\phi_0/4\pi\lambda^2)(m_1 B_X^2 + m_3 B_Z^2)^{1/2}\ln(H_{c2}\beta/B)\,. \tag{5}$$

The direction $\hat{\mathbf{Z}}$ coincides with the $\hat{\mathbf{c}}$ axis of the single crystal as is shown in Fig. 1; axis $\hat{\mathbf{X}}$ is chosen as the intersection of the $\hat{\mathbf{Z}}$-$\mathbf{B}$ plane with the basal plane normal to $\hat{\mathbf{Z}}$; thus, $B_Z = B\cos\theta$ and $B_X = B\sin\theta$ with θ being the angle between $\hat{\mathbf{c}}$ and $\mathbf{B}$. The "average" penetration depth λ is defined so that λ^2 is proportional to the average mass $M_{av} = (M_1^2 M_3)^{1/3}$, where the "effective masses" M_1 and M_3 are the components of M_{ik} along the principal crystal directions $\hat{\mathbf{X}}$ and $\hat{\mathbf{Z}}$, respectively.[3] For $Y_1Ba_2Cu_3O_7$ the ratio M_3/M_1, estimated from the ratio of H_{c2}'s in two principal crystal directions,[6,7] is in the range 25–90. Hereafter we use dimensionless masses $m_{ik} = M_{ik}/M_{av}$; we have then for the eigenvalues: $m_1^2 m_3 = 1$.

Differentiating F with respect to $\mathbf{B}$ one obtains the field

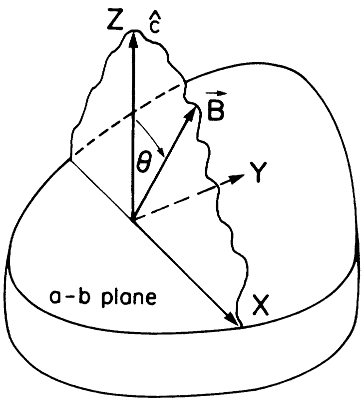

FIG. 1. A single-crystal grain in the applied field $H_a \gg H_{c1}$. The direction of vortices (**B**) almost coincides with $\mathbf{H}_a$ due to weak demagnetization effects. The a-b plane coincides with XY. The $\hat{\mathbf{X}}$ axis is the intersection of the a-b crystal plane with the plane $\hat{\mathbf{c}}$-$\mathbf{B}$.

H and the magnetization:

$$-M_Z = \frac{H_Z - B_Z}{4\pi} = M_0 \frac{m_3 \cos\theta}{\sqrt{m(\theta)}}, \quad -M_X = M_0 \frac{m_1 \sin\theta}{\sqrt{m(\theta)}}, \tag{6}$$

where

$$M_0 = \frac{\phi_0}{32\pi^2\lambda^2} \ln\frac{H_{c2}\beta}{H}, \quad m(\theta) = m_1 \sin^2\theta + m_3 \cos^2\theta. \tag{7}$$

In formulas for the small quantity **M**, the angle θ (between **B** and $\hat{\mathbf{c}}$) can be considered as the angle between the applied field $\mathbf{H}_a$ and $\hat{\mathbf{c}}$. Equations (6) and (7) are the constitutive relations for a uniaxial superconductor in the thermodynamic equilibrium; as such they cannot be applied to analyze magnetization curves when the latter are irreversible.

The component of **M** parallel to the applied field, M_p (which is measured routinely), is obtained readily from Eqs. (6) and (7):

$$M_p = M_Z \cos\theta + M_X \sin\theta = -M_0\sqrt{m(\theta)}. \tag{8}$$

It is worth noting that the component M_n normal to the applied field (which has recently been measured[8]) is not small compared to M_p:

$$M_n = M_X \cos\theta - M_Z \sin\theta = M_0 \frac{m_3 - m_1}{\sqrt{m(\theta)}} \sin\theta\cos\theta \tag{9}$$

(the directions $\hat{\mathbf{n}}$, $\hat{\mathbf{Y}}$, and **B** form a right-handed triad). The component M_n vanishes when the field is directed along one of the principal crystal directions $\theta = 0$ or $\pi/2$. However, the ratio $M_n/M_p = (m_3 - m_1)\sin\theta\cos\theta/m(\theta)$ reaches its maximum of $\sqrt{m_3/m_1}(m_3 - m_1)/2m_3$ at an angle θ_m such that $\tan^2\theta_m = m_3/m_1$. For $Y_1Ba_2Cu_3O_7$ the ratio M_n/M_p may be as large as 2.4–4.7 (depending on the value of m_3/m_1) at an angle $\theta_m \simeq 79°$–$84°$. It is interesting to note that the ratio M_n/M_p, evaluated here in the domain (1), is the same near H_{c2}.[9]

In particular, for the field applied along the $\hat{\mathbf{c}}$ axis ($\theta = 0$) and perpendicular to it ($\theta = \pi/2$) Eqs. (8) and (7) yield

$$-M(0) = \frac{\phi_0\sqrt{m_3}}{32\pi^2\lambda^2} \ln\frac{(H_{c2}\beta)_0}{H},$$
$$-M\left(\frac{\pi}{2}\right) = \frac{\phi_0\sqrt{m_1}}{32\pi^2\lambda^2} \ln\frac{(H_{c2}\beta)_{\pi/2}}{H}. \tag{10}$$

Thus, the magnetization of a single crystal in intermediate fields is given by Eqs. (6) and (7) in the crystal frame for any field direction, or by Eqs. (8) and (9) in the coordinate system aligned with the applied field for any crystal orientation. Because of weak demagnetization effects, the magnetic moment of the crystal is just $\mu = V_0\mathbf{M}$ where V_0 is the crystal volume. This allows one to obtain the magnetization of a sample made of randomly oriented crystalline grains of an average volume V_0 by a simple summation. The orientational distribution of the $\hat{\mathbf{c}}$ axes (given by the spherical angles θ and ϕ) with respect to the polar axis aligned with the applied field is given by $dN(\theta,\phi)/N = \sin\theta\, d\theta\, d\phi/2\pi$ with N being the number density of grains and $0 < \theta < \pi/2$. For each grain the component μ_n, normal to the field, is situated in the plane ($\hat{\mathbf{c}}$,$\mathbf{H}_a$); summation over all possible azimuthal orientations of $\hat{\mathbf{c}}$'s yields zero. Therefore, the magnetization of the polycrystal, M_{pc}, is directed along the applied field:

$$M_{pc} = N\int_0^{\pi/2} \mu_p(\theta)\sin\theta\, d\theta = \int_0^{\pi/2} M_p(\theta)\sin\theta\, d\theta, \tag{11}$$

where M_p is given in Eq. (8) and NV_0, the volume fraction of superconducting grains, is set equal to unity.

Actual integration in Eq. (11) is complicated by the angular dependence of $M_0 \propto \ln[H_{c2}(\theta)\beta]$, where the structural parameter β of the flux line lattice may be angular dependent as well. Fortunately, the slope $dM_{pc}/d(\ln H)$ can be evaluated readily:

$$\frac{dM_{pc}}{d\ln(H/H_0)} = \frac{\phi_0}{32\pi^2\lambda^2}\int_0^{\pi/2}\sqrt{m(\theta)}\sin\theta\, d\theta$$
$$= \frac{\phi_0}{64\pi^2\lambda^2}\sqrt{m_1}f(\gamma), \tag{12}$$

where H_0 is an arbitrary scaling field, and

$$\gamma^2 = m_3/m_1,$$
$$f(\gamma) = \gamma + (\gamma^2 - 1)^{-1/2}\ln[(\gamma^2-1)^{1/2} + \gamma]. \tag{13}$$

For the isotropic material $\gamma = m_1 = f/2 = 1$ and Eq. (12) yields the result which could have been obtained directly from Eq. (3). In the case of interest, $\gamma^2 \gg 1$, and to leading order $f = \gamma + O(1/\gamma)$. The normalization $m_1^2 m_3 = 1$ combined with the definition of γ yields $m_1 = \gamma^{-2/3}$ and $m_3 = \gamma^{4/3}$. Thus, the slope given in Eqs. (12) and (13) is expressed exclusively in terms of the averaged penetration depth λ and the anisotropy parameter γ.

The magnetization of a polycrystalline sample of $La_{1.85}Ba_{0.15}CuO_4$ measured by Finnemore and coworkers[10] is plotted in Fig. 2 as a function of H on a semi-

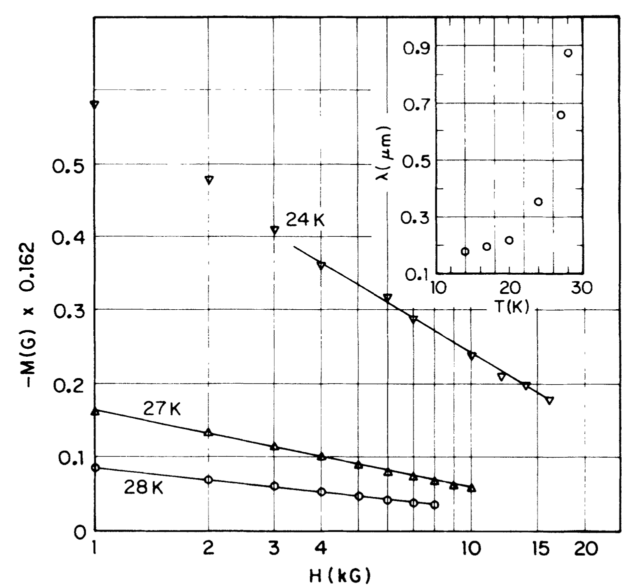

FIG. 2. The magnetization of a polycrystal LaSrCuO vs H. The factor 0.162 is to transform actual data (in milliemu) into the magnetization in gauss for the sample used. The inset shows the average penetration depth $\lambda(T)$, extracted from the $M(H)$ as is described in the text.

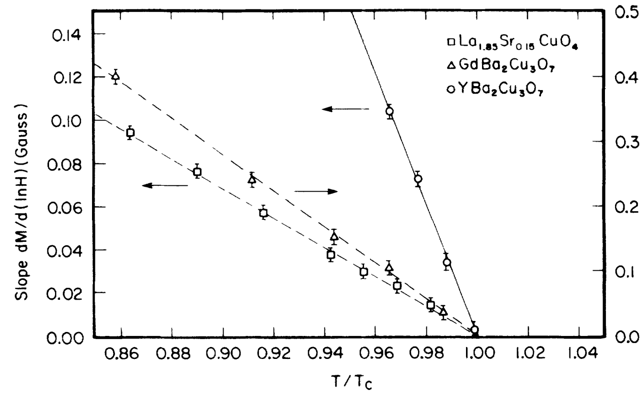

FIG. 3. The slopes $dM/d(\ln H)$ vs reduced temperature for three high-T_c compounds near their T_c's, extracted from the linear domains in M vs $\ln H$ data. For $YBa_2Cu_3O_7$ the data point closest to T_c belongs to $(T_c - T)/T_c \simeq 10^{-3}$.

log scale for a few values of temperature T in the field interval from 1 to 10 kG. One clearly sees the anticipated linear dependence of $M(\ln H)$. By evaluating the slope $dM/d(\ln H)$ from the graph one can estimate the average penetration depth λ with the help of Eqs. (12) and (13), provided the mass ratio γ^2 is known. Taking $\gamma = 6$ from the H_{c2} data obtained on a single-crystal $La_{1.9}Ba_{0.1}CuO_4$ in Ref. 11, one calculates λ shown in the inset of Fig. 2.[12] Note that the T dependence of λ obtained this way is qualitatively correct. The values of λ, however, may contain a factor (of the order unity) because the actual anisotropy parameter in the sample might have been different from that of Ref. 11. The estimate of λ is given just to demonstrate the method. Also, one should keep in mind that the T-independent anisotropy has been implied in the λ estimate.

Having found the average λ one can evaluate the penetration depth for any particular experimental situation. For example, the depth of a weak-field penetration (Meissner effect) into a slab with the $\hat{c}$ axis parallel to the slab plane depends on the field orientation. If $\mathbf{H}_a \parallel \hat{c}$, the actual penetration depth is $\lambda_{min} = \sqrt{m_1}\lambda = \gamma^{-1/3}\lambda$. For $\mathbf{H}_a \perp \hat{c}$ (and parallel to the slab surface), the effective depth is $\lambda_{max} = \sqrt{m_3}\lambda = \gamma^{2/3}\lambda$. If the field (still parallel to the slab plane) is inclined with respect to $\hat{c}$, its parallel and perpendicular components (with respect to $\hat{c}$) decay independently with λ_{min} and λ_{max}, respectively.

As has already been pointed out, the reversibility of $\mathbf{M}(H,T)$ is the necessary condition for the above theoretical arguments based on the free energy (5). It is certainly the case in the domain $T \to T_c$, where critical currents are small. In Fig. 3 we show the slopes $dM/d(\ln H)$ extracted from the linear parts of $M(\ln H)$ data for $La_{1.85}Sr_{0.15}CuO_4$, $YBa_2Cu_3O_7$, and $GdBa_2Cu_3O_7$ at temperatures close to their T_c's. Because $dM/d(\ln H) \propto \lambda^{-2}$, the Ginzburg-Landau theory yields $dM/d(\ln H) \propto (1 - T/T_c)$, the dependence clearly seen in Fig. 3.

It is worth noting that the model presented in this paper is based upon the mean-field theory. As such, it should break down in a narrow temperature domain $T_G < T < T_c$ near T_c, where the mean-field theory is no longer valid. Theoretical estimates of the Ginzburg parameter $G = (T_c - T_G)/T_c$ for $YBa_2Cu_3O_7$ vary from 10^{-1} (Ref. 13), via 10^{-2} (Ref. 14) and 10^{-3} (Ref. 15) to 10^{-5} (Ref. 16). The last data point for $YBa_2Cu_3O_7$ at Fig. 3, which still lies on the "mean-field straight line" belongs to $T = 92$ K, thus placing an upper limit upon the parameter G: $(92.1-92)/92.1 \simeq 10^{-3}$.

Two other possibilities are worth mentioning. If in the magnetization data taken on a single crystal the linear part of $M(\ln H)$ is well pronounced, one can, with the help of Eqs. (10), extract the anisotropy ratio m_3/m_1 by measuring the slopes $dM/d(\ln H)$ for two principal field orientations ($\theta = 0$ and $\pi/2$, at a fixed T) and by taking their ratio.

By extrapolation of the linear parts of $M(\ln H)$ to $M = 0$ one can estimate $H_{c2\beta}$, the quantity proportional to H_{c2}. However inaccurate such an estimate might be, it may provide an alternative to the direct determination of H_{c2} at low T's. Besides the difficulties associated with very large fields needed to reach H_{c2}, the direct measurement of the diamagnetic part of M (which $\to 0$ as $H \to H_{c2}$) might be complicated by other contributions to $\mathbf{M}$ unrelated to superconductivity.

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