

Dc Susceptibility in Type II Superconductors in Field-Cooled Process

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Dc susceptibility in oxide superconducting specimens in field-cooled process has been experimentally found to depend not only on applied dc magnetic field but also on size of specimens. The dc susceptibility is calculated using the critical state model in which the diamagnetism and the flux-pinning effect of superconductors are taken into account,. It is shown that a saturated value of the dc susceptibility at sufficiently low temperatures, i.e., the so-called Meissner fraction, decreases with increasing dc field and/or increasing specimen size.

1. Introduction

In the early stage of characterization of oxide superconducting specimens, the dc susceptibility at sufficiently low temperatures in field-cooled process has been adopted, under the recognition that its value gives a volume fraction of superconducting region in the specimens. However, it has been elucidated

[1,2] that the value of dc susceptibility varies with the external field even for a field sufficiently smaller than the lower critical field at low temperatures. The dc susceptibility has also been found [3] to depend on a size of specimens. These facts show that the dc susceptibility in the field-cooled process is not suitable for characterization of superconducting specimens.

The dc susceptibility directly corresponds to the magnetic flux distribution inside a specimen. There are two factors that determine the magnetic flux distribution: the intrinsic diamagnetism due to superconductivity that expels quantized magnetic fluxoids out of a specimen and the flux-pinning interaction that prevents motion of fluxoids. The magnetic flux distribution in type II superconductors has been known to be well described by the phenomenological critical state model. In this paper, the dc susceptibility in superconductors in the field-cooled process is treated using the critical state model. Dependences of the susceptibility on the dc magnetic field, the specimen size and the pinning strength in specimens are discussed.

2. Critical State Model

In the dc susceptibility measurement in the field-cooled process, the dc magnetic field is first applied to a superconducting specimen above the critical temperature and the measurement is started with decreasing temperature. For simplicity, we suppose that an infinite superconducting slab $0 \leq x \leq 2D$ is put in the parallel magnetic field of intensity H_e along the z -axis. Above the critical temperature, T_c , the magnetic flux density inside the superconductor is uniform and equal to $\mu_0 H_e$. From symmetry, we have only to consider a half of the superconducting slab, $0 \leq x \leq D$. When the temperature is reduced to $T = T_1 = T_c - \Delta T$ ($T > 0$), the slab becomes superconducting state and the diamagnetism appears. That is, equilibrium value of the magnetic flux density is reduced from $\mu_0 H_e$ to a certain value, which is denoted by $\mu_0 H_e + M(T_1)$ with $M(T_1) < 0$. Since the equilibrium magnetic flux density is reduced, excess of fluxoids are expelled out of the superconductor. If T_1 is larger than the irreversibility temperature under the magnetic field of H_e , $T_i(H_e)$, the critical current density is zero and no restrictional force acts on fluxoids. Hence, the flux distribution inside the superconductor is uniform and $B = \mu_0 H_e - M(T_1)$, as shown in Fig. 1(a). When the temperature is reduced to $T = T_n = T_i(H_e) - \Delta T$, the critical current density with a finite value, ΔJ_c , appears and the flux distribution depicted in Fig. 1(b) is

obtained. In the figure, the slope of the magnetic flux density is $\mu_0 \Delta J_c$. For further decreasing temperature, both the diamagnetism and the critical current density increase and we can expect the flux distribution given in Fig. 1(c). Repeating this procedure, finally we have the flux distribution shown in Fig. 1(d) at sufficiently low temperatures.

In the zero-field-cooled process, the flux distribution is much more easily obtained. According to elevating temperature, both the diamagnetism and the diamagnetic pinning interaction are weakened. Hence, the flux distribution is always determined by the diamagnetic magnetization and the critical current density at the measuring temperature alone, as shown in Fig. 2.

The flux distribution and the susceptibility at each temperature can be easily calculated numerically, if the temperature dependences of the diamagnetic magnetization, M , and the critical current density, J_c , are given. That is, if the mean flux density is denoted by $\langle B \rangle$, the relative susceptibility is given by

$$\chi = \frac{\langle B \rangle}{\mu_0 H_e} - 1. \quad (1)$$

According to Abrikosov [4], the diamagnetic magnetization for the magnetic field higher than the lower critical field $H_{c1}(T)$ is given by

$$M(T) = -\varepsilon \mu_0 [H_{c2}(T) - H_e], \quad (2)$$

where $\varepsilon = 1/1.16(2\kappa^2 - 1)$ and the temperature dependence of Ginzburg-Landau parameter κ is neglected. The temperature dependent upper critical field, $H_{c2}(T)$, is simply approximated as

$$H_{c2}(T) = H_{c2}(0) \left(1 - \frac{T}{T_c}\right). \quad (3)$$

Below the temperature, T_{c1} , at which the lower critical field coincides with the external field, we have

$$M(T) = -\mu_0 H_e, \quad (4)$$

Since the magnetic field dependence of M just above H_{c1} is complicated, we approximate it by a linear extrapolation of Eq.(2) from higher fields, as shown in Fig. 3. In this case ε should be modified from the original result

by Abrikosov, $1/1.16(2\kappa^2 - 1)$, in order to avoid a discrepancy at H_{c1} . For this purpose we use

$$\varepsilon = \frac{H_{c1}}{H_{c2} - H_{c1}}, \quad (5)$$

which is independent of temperature. Hence, ε is changed by a factor of $\log \kappa/1.16$. The characteristic temperature, T_{c1} , is expressed as

$$T_{c1} = \left[1 - \frac{(1 + \varepsilon)\delta}{\varepsilon} \right] T_c, \quad (6)$$

with

$$\delta = \frac{H_e}{H_{c2}(0)}. \quad (7)$$

Usually the external magnetic field in the dc susceptibility measurement is low and we can assume that the critical current density is rather independent of the magnetic field but depends only on the temperature. Here we assume as

$$J_c(H, T) = A \left(1 - \frac{T}{T_{c2}} \right)^n \quad (8)$$

where n is a parameter not smaller than 1 in usual and T_{c2} is a temperature at which J_c becomes zero. For simplicity, we assume that the irreversibility is alive up to the phase boundary, $B_{c2}(T)$. Thus, T_{c2} can be defined as a temperature at which the external dc field coincides with the upper critical field:

$$T_{c2} = (1 - \delta)T_c \quad (9)$$

Usually δ is much smaller than 1 and J_c can be regarded as almost constant with respect to magnetic field.

Examples of numerically calculated susceptibilities in field-cooled and zero-field cooled processes are represented in Fig. 4, where we have used $\mu_0 H_{c2}(0) = 100$ T, $T_c = 93$ K, $\varepsilon = 5.13 \times 10^{-4}$, $\mu_0 H_e = 1.0$ mT, $D = 1$ mm and $n = 1.8$. The value of A was varied from 1.0×10^8 to 1.0×10^{10} A/m². The dc susceptibility in the field-cooled process becomes saturated at sufficiently low temperatures. These results qualitatively agree with various experimental data[5]. Since we assumed that J_c has finite value up to T_{c2} as given in

Eq. (8), χ 's in the field-cooled and zero-field-cooled processes are different in the whole superconducting region. The difference in two χ 's becomes larger for larger A (stronger pinning force). If we replace T_{c2} in Eq. (8) by $T_i(H_e)$ and substitute $J_c = 0$ for $T \geq T_i$, the reversible region can be obtained, i.e., χ 's in the two processes meet on the same curve for $T_i \leq T \leq T_c$.

Although the above numerical calculation is simple, we cannot directly see the dependences of the saturated susceptibility at sufficiently low temperatures on the magnetic field, the specimen size and the pinning strength. Here we shall derive an analytic expression of susceptibility. The magnetic flux distribution near the surface region is given by

$$B(x) = \mu_0 H_e + M(T) + \mu_0 J_c(T)x, \quad (10)$$

which is represented by a linear part in Fig. 1(b) or 1(c). In the case of flux distribution shown in Fig. 1(d), the saturated susceptibility is calculated from an envelope, which is given by a locus of the penetration depth, x_0 , up to which the linear flux distribution shown by Eq. (10) holds. That is, the envelope is expressed by

$$\frac{\partial B(x_0)}{\partial T} = 0 \quad (11)$$

First, we treat the case of $T > T_{c1}$ where Eq. (2) is used for $M(T)$. Under the approximations given by Eqs. (2), (3), (8) and (9), Eq. (11) is written as

$$x_0 = \frac{\epsilon[H_{c2}(0) - H_e]}{An} \left[1 - \frac{T}{(1 - \delta)T_c} \right]^{1-n}, \quad (12)$$

The characteristic depth, x_0 , is indfinite at $T = T_{c2}$ and decreases monotonically with decreasing temperature for $n > 1$. The magnetic flux density at this depth is obtained from Eq. (10), which is still a function of the temperature, T . Elimination of T using Eq. (12) yields the envelope:

$$B(x) = \mu_0 H_e - (n - 1)\mu_0 \left[\frac{\epsilon H_{c2}(0)}{n} (1 - \delta) \right]^{n/(n-1)} \left(\frac{1}{Ax} \right)^{1/(n-1)} \quad (13)$$

for $x \geq x_0$.

It is to be noted that the above results are correct only for $n > 1$. For $n > 1$, from Eq. (12) we have $\frac{dx_0}{dT} > 0$. In this case the penetration depth of the linear flux distribution is not given by x_0 but by x'_0 as depicted in

Fig. 5. The region of the linear distribution given by Eq. (10) is developed according to decreasing temperature.

For the temperature below T_{c1} , the diamagnetic magnetization is constant and the boundary condition at the surface is fixed: $B(0) = 0$. Hence, the flux distribution at $T = T_{c1}$ is “frozen” even at lower temperature than T_{c1} . That is, the susceptibility is saturated for $T \leq T_{c1}$.

3. DC Susceptibility

In this section we shall theoretically derive the expressions of the dc susceptibility for various cases. If we represent the temperature at which x_0 coincides with D by T_0 , we have

$$T_0 = T_c(1 - \delta) \left\{ 1 - \left[\frac{\varepsilon H_{c2}(0)(1 - \delta)}{AnD} \right]^{1/(n-1)} \right\}. \quad (14)$$

First, we treat the case of $T_0 > T_{c1}$. This condition is written as

$$AnDH_e^{n-1} > \varepsilon^n H_{c2}^n(0)(1 - \delta)^n. \quad (15)$$

It can be seen that this condition is more easily satisfied for larger and/or more strongly pinned specimens. For $T_{c2} \geq T \geq T_0$, Eq. (10) represents the flux distribution in the whole sample and we obtain

$$\chi = \varepsilon - \frac{\varepsilon}{\delta} \left(1 - \frac{T}{T_c} \right) + \frac{AD}{2H_e} \left[1 - \frac{T}{(1 - \delta)T_c} \right]^n. \quad (16)$$

For $T_0 \geq T \geq T_{c1}$, the flux distribution is given by Eqs. (10) and (13) in the regions of $0 \leq x \leq x_0$ and $x_0 \leq x \leq D$, respectively. The susceptibility is

$$\chi = -\frac{\varepsilon^2 H_{c2}^2(0)(1 - \delta)^2}{2n(2 - n)DAH_e} \left[1 - \frac{T}{(1 - \delta)T_c} \right]^{2-n} + \frac{(n - 1)^2}{(2 - n)(AD)^{1/(n-1)}H_e} \left[\frac{\varepsilon H_{c2}(0)}{n} (1 - \delta) \right]^{n/(n-1)}. \quad (17)$$

For $T \leq T_{c1}$, the susceptibility is saturated t

$$\begin{aligned} \chi_s = & -\frac{\varepsilon^n H_{c2}^n(0)(1 - \delta)^n}{2n(2 - n)DAH_e^{n-1}} \\ & + \frac{(n - 1)^2}{(2 - n)(AD)^{1/(n-1)}H_e} \left[\frac{\varepsilon H_{c2}(0)}{n} (1 - \delta) \right]^{n/(n-1)}. \end{aligned} \quad (18)$$

It should be noted that Eqs. (17) and (18) hold for $n \neq 2$. In the case of $n = 2$, it is easily shown that Eqs. (17) and (18) are replaced by

$$\chi = -\frac{\varepsilon^2 H_{c2}^2(0)(1-\delta)^2}{4ADH_e} \left[\frac{3}{2} + \log \left\{ \frac{2AD}{\varepsilon H_{c2}(0)(1-\delta)} \left[1 - \frac{T}{(1-\delta)T_c} \right] \right\} \right]; T_0 \geq T \geq T_{c1}, \quad (19)$$

and

$$\chi = \chi_s = -\frac{\varepsilon^2 H_{c2}^2(0)(1-\delta)^2}{4ADH_e} \left[\frac{3}{2} + \log \frac{2ADH_e}{\varepsilon^2 H_{c2}^2(0)(1-\delta)^2} \right]; \quad T \geq T_{c1}, \quad (20)$$

respectively.

In the case of $T_0 < T_{c1}$, i.e., if Eq. (15) is not satisfied, the flux distribution is given by Eq. (10) alone until T is reduced to T_{c1} . That is, χ is given by Eq. (16) for $T_{c2} \geq T \geq T_{c1}$. For $T < T_{c1}$, χ is saturated to

$$\chi_s = -1 + \frac{ADH_e^{n-1}}{2[\varepsilon H_{c2}(0)(1-\delta)]^n}. \quad (21)$$

The susceptibility for the case of $n \leq 1$ can also be obtained similarly. The results are shown in Appendix.

In the zero-field-cooled process the magnetic flux distribution varies as shown in Fig. 2. The distribution near the surface region obeys

$$B(x) = \mu_0 H_e + M(T) - \mu_0 J_c(T)x. \quad (22)$$

The point, x_0'' , at which B reaches zero given by

$$x_0'' = \frac{H_e}{A} \left[1 - \frac{T}{(1-\delta)T_c} \right]^{-n} - \frac{\varepsilon H_{c2}(0)(1-\delta)}{A} \left[1 - \frac{T}{(1-\delta)T_c} \right]^{1-n}. \quad (23)$$

This point appears only for $T \geq T_{c1}$ and its value increases monotonically with increasing temperature and becomes infinite at $T = T_{c2}$. If we represent the temperature at which x_0'' coincides with D , T_0'' can be obtained only numerically. For $T \leq T_{c1}$, $B = 0$ throughout the specimen and we have

$\chi = 0$. For $T_{c1} \leq T \leq T_0''$, the flux distribution is given by Eq. (22) and $B(x) = 0$ in the regions of $0 \leq x \leq x_0''$ and $x_0'' \leq x \leq D$, respectively. The susceptibility in this case is

$$\begin{aligned} \chi = & -1 + \frac{H_e}{2DA} \left[1 - \frac{T}{(1-\delta)T_c} \right]^{-n} - \frac{\varepsilon H_{c2}(0)(1-\delta)}{DA} \left[1 - \frac{T}{(1-\delta)T_c} \right]^{1-n} \\ & + \frac{\varepsilon^2 H_{c2}^2 (1-\delta)^2}{2DAH_e} \left[1 - \frac{T}{(1-\delta)T_c} \right]^{2-n}. \end{aligned} \quad (24)$$

For $T \geq T_0''$, the flux distribution is expressed by Eq. (22) in the whole sample and we have

$$\chi = -\frac{\varepsilon H_{c2}(0)(1-\delta)}{H_e} \left[1 - \frac{T}{(1-\delta)T_c} \right] - \frac{AD}{2H_e} \left[1 - \frac{T}{(1-\delta)T_c} \right]^n. \quad (25)$$

These results on the susceptibility in the zero-field-cooled process hold also for $0 < n \leq 1$.

4. Discussion

Here we discuss the teoretical result obtained from the critical state model with respect to dependences of the saturated susceptibility on external magnetic field, sample size and pinning strength.

The saturated dc susceptibility, χ_s , depends on the external magnetic field as shown in Fig.4. Figrure 6 represents the observed χ_s for single crystal La-Sr-Cu-O ($D=0.1$ mm) vs the magnetic field along the a-axis [3, 4]. The solid line is the teoretical result with $T_c = 35$ K, $\mu_0 H_{c2}(0) = 27.3$ T, $\varepsilon = 5.13 \times 10^{-4}$, $A = 3.2 \times 10^{10}$ A/m² and $n = 1.8$. Agreement is satisfactory in spite of various asumptions in the teoretical calculation. It should be noted here that such a large value of A cannot be obtained from a hysteresees in dc magnetization curve by using Bean's model for a large sample. That is, the field necessary for a complete penetration of the flux is fairly high for a large sample and the critical current density varies largely inside the sample. In this case, a fit of the dc magnetization hysteresis to the model in which the field dependence of the critical current density is taken into account is better. For $n > 1$, the depth of linear flux distribution, x_0 , is very large near T_{c2} . This means that the magnetic flux expelled from the sample is relatively larger when a variation in $B(0)$ from $\mu_0 H_e$ is smaller. Hence,

it can be qualitatively explained that the saturated dc susceptibility, $-\chi_s$ is larger under a smaller magnetic field.

Figure 7 depicts the saturated dc susceptibility vs magnetic field for various sample size. Figure 8 is a replot of Fig.7 and shows the sample size dependence of $-\chi_s$. The value of $-\chi_s$ becomes smaller for a larger sample. Observed $-\chi_s$'s in a single crystal and powders of Bi-Sr-Ca-Cu-O [6] are shown in Fig.9. This result qualitatively agrees with the predicted sample size dependence by the critical state model. Since the magnetic flux is expelled from the sample surface, the more flux is remained inside the larger sample, resulting in smaller $-\chi_s$.

The dependence of $-\chi_s$ on the pinning strength, A , is shown in Fig.10. The value of $-\chi_s$ becomes smaller according to increasing pinning strength. This is natural, since stronger pinning interaction traps more magnetic flux inside the sample.

As shown in this paper, the value of dc susceptibility in the field-cooled process at sufficiently low temperatures depends on strongly on external magnetic field, sample size and pinning strength, although the volume fraction of superconducting region, i.e., the true Meissner fraction is assumed to be 100 %. This means that $-\chi_s$ does not give correct value of the Meissner fraction and characterization of samples by measurements of $-\chi_s$ is not justified. In particular, underestimate of a quality of sample is significant for a large and strongly pinned sample. Figure 11 shows the observed susceptibilities of a melt-processed Y-Ba-Cu-O in the field-cooled and zero-field-cooled processes at $\mu_0 H_e = 5$ mT [5]. It is well known that this material has a high quality and strong pinning force even for a sample with large size [7]. In general, samples with large J_c 's and moderate sizes are needed from the viewpoint of application. In this sense, a sample with smaller $-\chi_s$ in the field-cooled process is rather of better quality.

It is shown that the behavior of trapped magnetic flux in a superconductor can be qualitatively described by the simple critical state model. In a strict sense, however, a long-range diamagnetic force on trapped fluxoids which has been observed in low T_c superconductors [8] might be involved below H_{c1} in high T_c superconductors, too. If it is so, fluxoids would be continuously expelled from the sample according to decreasing temperature even below T_{c1} . This is not clear at this moment and further detailed investigation is necessary in order to exactly understand the magnetic behavior in high T_c superconductors.

Appendix

(i) $n < 1$

The flux distribution in this case changes as shown in Fig. 5 with decreasing temperature. The position, x'_0 , at which B reaches $\mu_0 H_e$ is expressed as

$$x'_0 = \frac{\varepsilon H_{c2}(0)(1-\delta)}{A} \left[1 - \frac{T}{(1-\delta)T_c} \right]^{1-n}. \quad (\text{A-1})$$

If T'_0 denotes the temperature at which x'_0 is equal to D , we have

$$T'_0 = T_c(1-\delta) \left\{ 1 - \left[\frac{\varepsilon H_{c2}(0)(1-\delta)}{AD} \right]^{1/(n-1)} \right\}. \quad (\text{A-2})$$

In the case of $T'_0 > T_{c1}$, i.e.,

$$ADH_e^{n-1} < [\varepsilon H_{c2}(0)(1-\delta)]^n, \quad (\text{A-3})$$

the susceptibility is given by

$$\chi = -\frac{\varepsilon^2 H_{c2}^2(0)(1-\delta)^2}{2ADH_e} \left[1 - \frac{T}{(1-\delta)T_c} \right]^{2-n} \quad (\text{A-4})$$

for $T'_0 \leq T \leq T_{c2}$, Eq. (16) for $T_{c1} \leq T \leq T'_0$ and Eq. (21) for $T \leq T_{c1}$. If $T'_0 > T_{c1}$, the susceptibility is given by Eq. (A-4) for $T_{c1} \leq T \leq T_{c2}$ and

$$\chi = \chi_s = -\frac{[\varepsilon H_{c2}(0)(1-\delta)]^n}{2ADH_e^{n-1}} \quad (\text{A-5})$$

for $T \leq T_{c1}$.

(ii) $n = 1$

Equation (A-1) for the characteristic depth, x'_0 , is useful also in this case. Now x'_0 is independent of the temperature. If $x'_0 > D$, we have Eq. (16) for $T_{c2} \geq T \geq T_{c1}$ and Eq. (21) for $T \leq T_{c1}$. If $x'_0 < D$, we have Eq. (A-4) for $T_{c2} \geq T \geq T_{c1}$ and Eq. (A-5) for $T \leq T_{c1}$.

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Figure Captions

Fig. 1. Schematic illustrations of variation of flux distribution inside a superconductor in field-cooled process: (a) Flux distribution is uniform for $T \geq T_i$. (b) When T is decreased from T_i by ΔT , irreversibility appears and some flux is prevented from going out. (c) Diamagnetism and pinning effect become stronger with decreasing temperature. (d) Flux distribution at sufficiently low T .

Fig. 2. Variation of flux distribution in zero-field-cooled process.

Fig. 3. Simplified magnetic field dependence of intrinsic magnetization.

- Fig. 4.** Examples of numerically calculated susceptibilities in field-cooled and zero-field-cooled processes for samples with various pinning strength. Used parameters are $T_c=93$ K, $\mu_0 H_{c2}(0)=100$ T, $\varepsilon=5.13 \times 10^{-4}$, $n=1.8$, $D=1$ mm, $\mu_0 H_e=1$ mT and $A=1.0 \times 10^8$, 1.0×10^9 and 1.0×10^{10} A/m².
- Fig. 5.** Variation of flux distribution in field-cooled process for $n < 1$.
- Fig. 6.** Saturated susceptibility vs magnetic field. Symbols show experimental result on La-Sr-Cu-O single crystal [6] and a solid line represents the theoretical result.
- Fig. 7.** Saturated susceptibility vs magnetic field for various sample sizes. Assumed parameters are $T_c=93$ K, $\mu_0 H_{c2}(0)=100$ T, $\varepsilon = 5.13 \times 10^{-4}$, $A=1.0 \times 10^{10}$ A/m² and $n=1.8$.
- Fig. 8.** Sample-size dependence of saturated susceptibility at $\mu_0 H_e=1.0$ mT. This is a reprot of Fig. 7 and the other parameters assumed are the same.
- Fig. 9.** Observed saturated susceptibilities in a single crystal and powders of Bi-Sr-Ca-Cu-O [6].
- Fig. 10.** Saturated susceptibility vs magnetic field for various pinning strength. Assumed parameters are $T_c=93$ K, $\mu_0 H_{c2}(0)=100$ T, $\varepsilon = 5.13 \times 10^{-4}$, $n=1.8$ and $D=1$ mm.
- Fig. 11.** Susceptibilities of a melt-processed Y-Ba-Cu-O in field-cooled and zero-field-cooled processes at $\mu_0 H_e=5$ mT.