Penetration depth and anisotropy in MgB$_2$

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The penetration depth λ of MgB$_2$ was deduced from both the ac susceptibility χ and the magnetization $M(H)$ of sorted powders. The good agreement between the two sets of data without geometric correction for the grain orientation suggests that MgB$_2$ is an isotropic superconductor.

Great interest has been raised recently by the discovery[1] of MgB$_2$ with T_c about 40 K. In its normal state, the compound appears to be a metal with a low dc resistivity,[2] Hall coefficient,[3] and thermoelectric power[4] dominated by hole carriers. The estimated long mean free path[2] implies that the electrical transport could well be isotropic in spite of its layerlike crystalline structure, in agreement with the band-structure calculations.[5] Below a transition temperature of $T_c \approx 40$ K, the compound seems to be a phonon-mediated BCS superconductor, as suggested by the large isotopic effect[6] and the large negative pressure effect[4] on T_c. The upper critical field, $H_{c2}(T)$, has been directly measured above 20 K with a linear (or even an upward curvature) T dependence.[7] The lower critical field was either indirectly estimated based on the thermodynamics field $H_c(T)$ above 34 K or calculated based on the nonlinearity in $M(H)$ at low temperatures.[7,8] The coherence length ξ_0 (0 K), penetration depth λ (0 K), and Ginzburg-Landau parameter κ were consequentially estimated to be 5.2 nm, 125–140 nm, and 26, respectively. All these calculations, however, were based on the assumption that MgB$_2$ can be treated as isotropic. Otherwise, a geometric factor up to 2 would be needed to correct the random grain orientation and to convert M_r to H_c.

To estimate the anisotropy, λ of MgB$_2$ was directly measured using both ac susceptibility and the nonlinearity in the $M(H)$ of powder samples. Although the two procedures involve the anisotropy of λ in very different ways, the λ(5 K)'s deduced from these procedures are in good agreement without geometric corrections for superconducting anisotropy. The results therefore suggest that the anisotropy of MgB$_2$ is very small.

Ceramic MgB$_2$ samples were prepared using the solid-state reaction method.[6] Small Mg chips (99.8% pure) and B powder (99.7% pure) with a stoichiometry of Mg:B=1:2 were sealed inside a Ta tube under an Ar atomosphere. The sealed Ta ampoule was then enclosed in a quartz tube. The assembly was heated slowly up to 950 °C and was kept at this temperature for 2 h, followed by furnace cooling. The structure was determined by x-ray powder diffraction (XRD) using a Rigaku DMAX-IIIB diffractometer. Powder samples were prepared by sorting the pulverized powder using either sieves or the method of descending speed of the particles in acetone. No grain alignment was attempted. The grain morphology as well as the particle sizes of the powders were measured using a JEOL JSM 6400 scanning electron microscope (SEM). Magnetizations were measured in a Quantum-Design 5 T superconducting quantum interference device

magnetometer with an ac attachment. The ac susceptibility was measured under a fixed frequency $f = 413.1$ Hz and an amplitude of 3 Oe.

The XRD pattern can be indexed as a hexagonal cell with lattice parameters $a=3.08$ Å and $c=3.52$ Å. A sharp superconducting transition was observed in both resistivity and ac susceptibility with $T_c \approx 38$ K.[4]

The transition temperature in different fields, $T_c(H)$, was determined from both the dc magnetization and the ac susceptibility measurements, which should be equivalent to a local resistivity measurement, with a dc bias of 0–5 T. The transition width in the ac susceptibility is only slightly broadened under fields, i.e., from ≈ 1 K at the bias field of $H_{dc} = 0$ T to ≈ 5 K at 5 T (inset, Fig. 1). We attribute this to the flux movement under fields and take the onset temperature as $T_c(H)$ (Fig. 1). The data from the two methods are reasonably consistent with a slope $dH_{c2}/dT \approx 0.4 \pm 0.05$ T/K near 38 K, in agreement with the results obtained by Finnemore *et al.*[7]

The lower critical field was measured in powder samples with a particle size <2 μm to avoid the complications caused by intergrain coupling.[9] No systematic variation be-
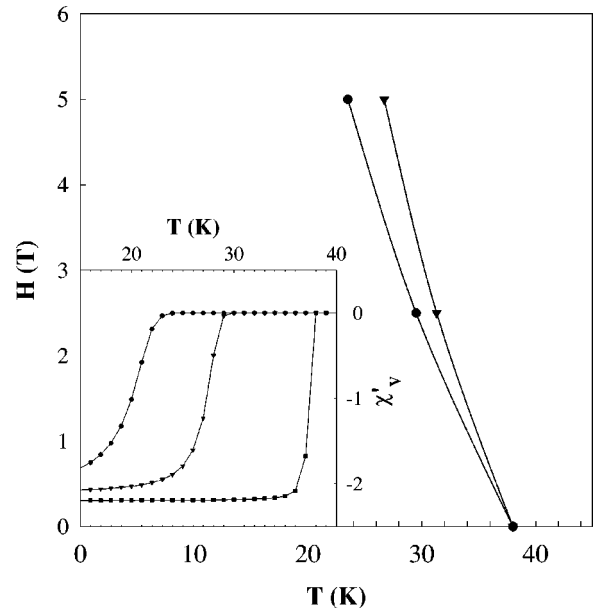

FIG. 1. H_{c2} of a MgB$_2$ ceramic sample. ▼: from the dc magnetization; ●: from the ac susceptibility with a dc bias H. Inset: the ac susceptibility at $H=$ ■: 0 T; ▼: 2.5 T; ●: 5 T.

FIG. 2. SEM photo of the powder sample.

FIG. 3. $1/\lambda^2(T)$ of a MgB$_2$ powder sample. $\bigcirc$: data; solid line: fit as $\propto [1 - (T/39.4)^{2.7}]$. Inset: the particle-size distribution of the powder.
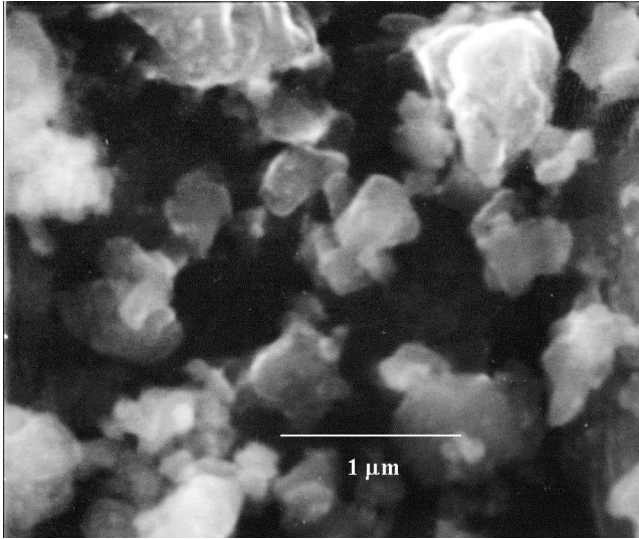
tween the deduced $\lambda(T)$ and the particle sizes was observed. This demonstrates that the grain-boundary effect in the sample is small, a fact that is also supported by direct SEM observation of the powders.

It is known that the ac susceptibility χ of a superconducting sphere of diameter d is $\Phi(d,\lambda) = -3/(8\pi)[1 - 6(\lambda/d)\coth(d/2\lambda) + 12(\lambda/d)^2]$, which reduces to $\propto (d/\lambda)^2$ when $d < 2\lambda$. Methods have been previously developed to deduce λ from the χ of magnetically aligned powders by solving the equation $\chi = \Phi(d,\lambda) \approx -0.002(d/\lambda)^2$ in the large-λ limit.[10] However, the λ so deduced will be sensitive to the uncertainty of χ (due to either the demagnetization factor or the superconducting volume fraction) if $d \gg 2\lambda$ as in unsorted powders, which may include particles as large as 3 μm.[10] In fact, the calculated $\partial \ln \lambda / \partial \ln \chi$ varies with d/λ only moderately below $d/\lambda = 4$, i.e., from -0.5 to -0.7, but changes rapidly for larger d/λ. For example, $\partial \ln \lambda / \partial \ln \chi$ is -3 at $d/\lambda = 20$, and a 30% uncertainty of χ will lead to a λ anywhere between 0 and $10d$. A $d/\lambda < 5$ is needed to obtain a 20% accuracy with an estimated 30% uncertainty in χ. The technique can be improved by using sorted powders, which have a smaller d and a narrower size distribution.[11] The powder obtained from pulverizing ceramic was thoroughly mixed with acetone in a 10-ml beaker. The particles was then sorted according to the time needed for them to reach the bottom of the beaker. The sample discussed here was collection of particles deposited between 1–2 h. Our SEM observation suggested that 99% or more particles having a size between 0.1 and 2 μm (Fig. 2). It should also be noted that the proposed method of calculating the effective grain size $d = \sqrt{(\Sigma d_i^5)/(\Sigma d_j^3)}$ (where d_i is the diameter of individual grains) may also be questionable if $d > 2\lambda$.[10] As will be shown below, a 30% error may be caused by the approximation alone. A regression procedure therefore was adopted. A λ_{raw} corresponding to the $d_{raw} = \sqrt{(\Sigma d_i^5)/(\Sigma d_j^3)}$ was used as the initial value. The d was then refined regressively as $[\Sigma \Phi(\lambda, d_i) d_i^4]/[\Sigma \Phi(\lambda, d_i) d_i^3]$. The convergence is very fast. It should be noted that the effective d depends on λ, i.e.,
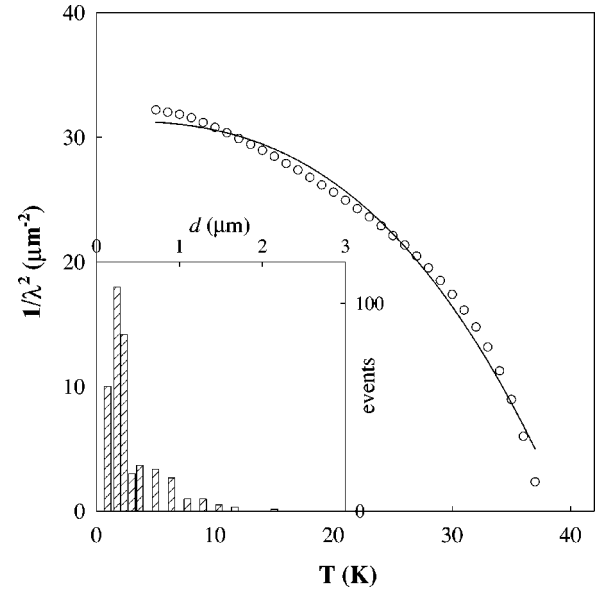
the correction varies with T and may change the T dependence of λ. Our tests on YBa$_2$Cu$_3$O$_{7-\delta}$ powders demonstrated that the uncertainty of the λ so deduced is within 10–20 % of the published data if d/λ is 3 or smaller.[11]

The SEM photo of a powder sample is shown in Fig. 2. Its d distribution is shown in the inset of Fig. 3. A $d \approx 0.88$ μm was obtained with $d_{raw} = 1.23$ μm. The superfluid density $1/\lambda^2(T)$ was calculated assuming an isotropic superconductivity, i.e., without corrections for the random grain orientation (Fig. 3). The $1/\lambda^2(T)$ observed roughly follows a T dependency of $[1 - (T/39.4)^{2.7}]$. It should be noted that a deviation from the fit is clear below 10 K. We are hesitate, however, to draw any conclusion about the deviation since weak links cannot be conclusively excluded at this stage. The extrapolated $\lambda(0) \approx 180$ nm is slightly longer than that found by Finnemore et al.[7]

The lower critical field H_{c1} was also deduced as the field, where the linear $M-H$ correlation begins to be violated, from the magnetizations of the same powder sample in an H-increase branch at 5 K (inset, Fig. 4). Several technical difficulties in this method have been previously discussed: for instance, the intergrain coupling that may cause nonlinearity far below the intragrain H_{C1}; the surface pinning that can make the H_{C1} value observed higher; sharp local edges, i.e., strong local demagnetizing fields, that can lead to a lower one; and the experimental resolution of the nonlinearity. Several precautions have been taken. The powder sample used here has a particle size far smaller than the average grain size, which should eliminate the effect of the intergrain coupling, as suggested by the smooth and flat χ observed below T_c. To improve the sensitivity of nonlinearity, the $M(H, 5 \text{ K})$ below 50 Oe was fit as a linear function of H using a standard least-square procedure. The deviation $\Delta M (\approx 0.002$ emu/cm^3 below 50 Oe) from the linear fit is
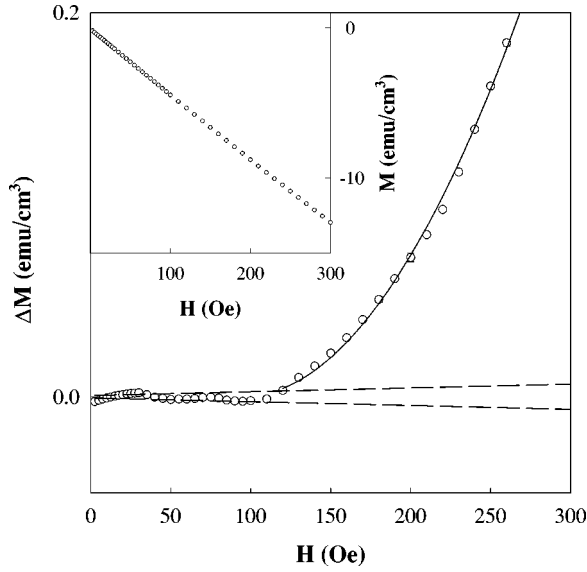
FIG. 4. The deviation ΔM from the linear extrapolation at 5 K. $\bigcirc$: data; dashed lines: the uncertainty bands of the linear fit; solid line: a fit of $(H-H_0)^{1.8}$. Inset: $M(H)$ at 5 K.

FIG. 5. The ac susceptibility with a dc bias H at 5 K.

comparable with the experimental uncertainties in both M and H, demonstrating the negligible effect of the residual granularity. The difference between the data and the extrapolated linear fit above 50 Oe was then calculated. The uncertainty associated with the linear fitting was marked as dashed lines in Fig. 4. To further avoid the interferences from the sharp edges of the particles, the deviation at large fields was empirically fit as $a \cdot (H-H_0)^{1.8}$ with both a and H_0 as the free parameters (Fig. 4). We justify the fit by pointing out that the magnetization of a superconductor partially penetrated by external fields will vary as the square of the thickness penetrated, i.e., $H-H_0$ in the Bean model. It should be pointed out that the value of H_0 is not very sensitive to the index chosen. A linear fit below 200 Oe leads to only 15% change. Surface pinning is usually negligible in randomly shaped grains, and typically can only make the H_{c1} observed smaller. Following the procedure, an $H_{c1}=H_0/(1-g)$ ≈ 130 Oe was obtained, where $g=1/3$ is the demagnetization factor of a sphere. This value consequently leads to a λ of 203 nm, in good agreement with that from the ac χ within the uncertainty of the techniques. No corrections have been made to consider random grain orientations.

To further verify the result, the ac susceptibility of the same powder sample was measured at 5 K with a dc bias between 0 and 200 Oe (Fig. 5). A change of the slope was observed around $H_0 \approx 110$ Oe, and the H_{c1} was estimated as ≈ 160 Oe. Similar measurements have been done in several different samples and the results appear to be independent of the particle sizes.

The deduced λ is slightly longer than the 140 nm (Ref. 7) and the ≈ 130 nm (Ref. 8) previously reported. The exact reason for the disagreement is not clear to us at this moment. However, the λ measured here using three different methods on the same sample are self-consistent within the estimated experimental uncertainty of $\pm 20\%$.

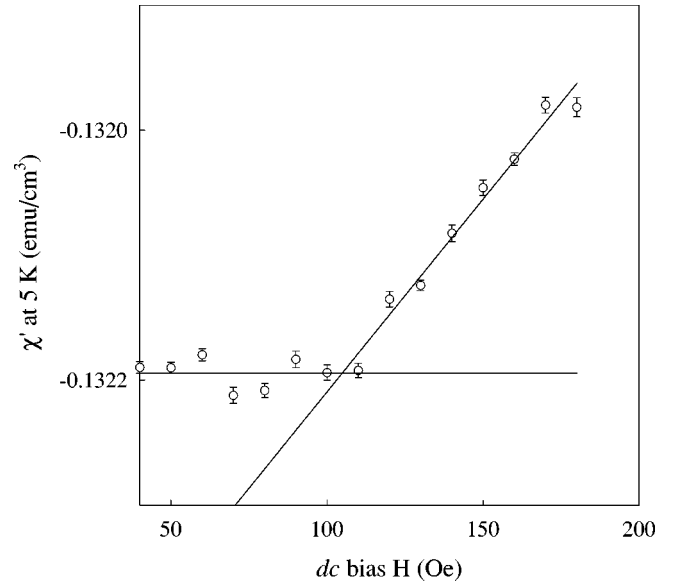It is interesting to note the agreement between the H_{c1}

from χ and $M(H)$. In a highly anisotropic layered superconductor, cuprates for example, the observed χ will only come from the supercurrents in the layers. The χ observed therefore should be assumed to be $\int \cos^2 \theta \sin \theta d\theta / \int \sin \theta d\theta \approx 1/3$ of the $-3/(8\pi)[1-6(\lambda/d)\coth(d/2\lambda)+12(\lambda/d)^2]$. The deduced λ will be 1.7 times longer if no geometric correction has been made. In general, the $1/\lambda^2$ deduced from non-grain-aligned powder should be $1/3\lambda_{ab}^2 + 2/3\lambda_c^2 = (1/3+2/3\gamma)/\lambda_{ab}^2$ in layered superconductors, where λ_{ab}, λ_c, and γ are the penetration depths in and out of the layers, and the anisotropy, respectively. The H_{c1} deduced from the nonlinearity of $M-H$, on the other hand, can be even larger since the effective field perpendicular to the local layers is only a fraction, i.e., $\cos \theta$, of the external field. The good agreement observed here therefore strongly suggests that MgB$_2$ is an isotropic superconductor. The estimated γ should be smaller than 1.5 assuming an experimental uncertainty of $\pm 20\%$ in our λ calculation. This γ is far smaller than that of 10–1000 observed in various cuprates, and should be regarded as essentially isotropic.

In the above data analysis, a spherical shape was assumed. The relative change of the demagnetization factor is $\Delta r/3r$ in a slightly deformed ellipsoid with radii $r+\Delta r$, $r+\Delta r/2$, and $r+\Delta r/2$. A simple calculation shows that the correction of the H_{c1} will be $-0.25(\Delta r/r)^2$ in the ac χ method,[12] but $\Delta r/3r$ in the nonlinearity method. An average length ratio $(r+\Delta r)/(r-\Delta r)$ between 0.5 and 2 (i.e., $|\Delta r/3r|=1/3$) therefore may not significantly change the above conclusion. The condition seems to be satisfied (Fig. 2).

This conclusion is in agreement with the band-structure calculation, the extremely long mean free path, the long coherence length, and the small grain-boundary effect on the supercurrents reported.

In summary, the penetration depth $\lambda(T)$ of MgB$_2$ was

deduced from both the ac susceptibility χ of powders and the nonlinearity of the $M-H$ in the H-increase branch. The good agreement between the two methods suggests that MgB_2 is an isotropic superconductor.

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