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High temporal stability of supercurrents in MgB$_2$ materials

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Abstract
Fine grained polycrystalline samples of MgB$_2$ superconductor were
synthesized from the elements to contain < 5 % of impurity phases,
according to x-ray powder diffractometry. The superconductive transition
was sharp with a midpoint $T_c = 38.5$ K. The magnetization in the vortex
state was studied as a function of applied field H, temperature T and time t.
From the equilibrium magnetization, the London penetration depth was
obtained. The supercurrent density $J(T, H, t)$ in the vortex state (derived
from the irreversible magnetization) decreases approximately linearly with
T, in contrast to the quasi-exponential fall-off in high-T_c superconductors.
The current is highly stable in time, with normalized decay rates
$S = -\,\mathrm{d}\ln(J)/\mathrm{d}\ln(t)$ well below those in high-T_c materials. These results
are compared with those of other superconductors.

1. Introduction

The recent discovery [1] of superconductivity near 40 K in
MgB$_2$ has stimulated intense interest in this compound. An
observation of a large boron-isotope effect [2] points to a
central importance for electron–phonon coupling. Studies of
the conduction of critical currents [3, 4] in polycrystalline
materials in large magnetic fields are encouraging: the grains
appear to be well linked, in stark contrast with high-T_c
cuprate superconductors (HTSC) with their vexing problem
of weak-linkage between grains. Another prominent feature
of the HTSCs is a rapid decay of (nearly) critical currents
in the mixed-state, which arises from easy movement of
vortices ('giant flux creep') that are depinned either by thermal
activation or by quantum tunnelling [5]. Thus, it is important
to investigate the temporal stability of supercurrents in this
new material with its markedly different structure and simple
composition.

In this work, we investigate several mixed-state properties
of an array of isolated MgB$_2$ grains. Thus, we obtain the
London penetration depth, which we use to establish the basic
scale of current density—the depairing current density. This
we compare with the experimentally realized current density
and explore its dependence on temperature and magnetic field.
Finally, the normalized decay rate is presented, then compared

with the rates in several other superconductors. We find that
for a substantial range of field and temperature, the mixed-state
supercurrent density is highly stable in time, in contrast with
most HTSC materials.

2. Experimental aspects

The materials were prepared using high purity elemental
starting materials. Fine grained polycrystalline samples of
superconductor were synthesized by reaction of the elements
at 890°C for 2 h in a crimped Ta tube, contained within an
evacuated quartz ampoule. The resulting materials contained
< 5 % of impurity phases according to x-ray diffraction. In
the form of a porous sintered pellet, the material exhibited
a sharp superconductive transition with a midpoint $T_c =
38.5$ K. The shielding was complete, as measured in an applied
field of 0.4 mT. For studies in the mixed state, the sample
was powdered and dispersed in a clear epoxy (Stycast 1267)
with a volume fraction of $\sim$2 %. This procedure reduces
the magnitude of the irreversible magnetization relative to
the equilibrium component, as this ratio is proportional to
$\langle d^4\rangle/\langle d^3\rangle \sim d$, where d is a particle's transverse dimension.
The average particle dimension was determined by optical
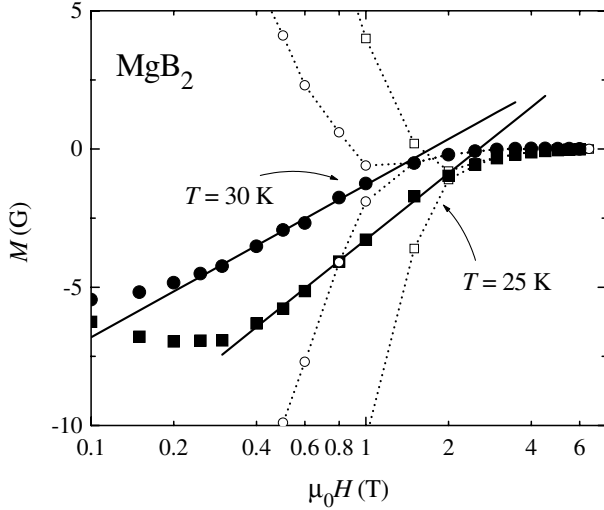measurement of particles contained in thin sections cut from

Figure 1. The magnetization M of isolated particles of MgB$_2$ at temperatures of 25 K (squares) and 30 K (circles), plotted versus applied magnetic field on a log scale. Open symbols show experimental measurements made in increasing and decreasing fields, while closed symbols denote the average, equilibrium magnetization. Solid curves show fits to standard London theory, equation (1).

Figure 2. The London penetration depth plotted as $1/\lambda^2$ versus temperature. The dashed curve illustrates a Ginzburg–Landau dependence near T_c, while the solid curve is a fit to BCS theory in the clean limit.

3. Experimental results and discussion

The magnetization of small, isolated grains of MgB$_2$ was studied in the superconductive state, as described above. Two examples of these data are shown in figure 1, which plots M at temperatures of 25 and 30 K versus applied field $\mu_0 H$ on a log scale. The open symbols show measured values and illustrate the hysteretic magnetization in the mixed state. First we consider the equilibrium magnetization obtained by averaging the up-field and down-field values. The results are denoted by the solid symbols in figure 1. Now, in ordinary London theory, M_{eq} has a simple logarithmic field dependence, [6] for fields $H_{c1} \ll H \ll H_{c2}$, with

$$M(H, T) = -\varphi_0 \left(32\pi^2\lambda^2(T)\right)^{-1} \times \ln(\eta H_{c2}/B) \quad (1)$$

Here φ_0 is the flux quantum, λ is the London penetration depth, and η is a constant of order unity. The semi-log plot of M versus $\mu_0 H$ in figure 1 shows that this widely used relation provides a good description of the data for a significant field range, as shown by the solid curves that are fitted to equation (1). From the slopes of these and similar fits, we obtain values for $\lambda(T)$.

The results for the temperature dependence of the London penetration depth are shown in figure 2 as a plot of $1/\lambda^2(T)$. According to Ginzburg–Landau theory, one has $1/\lambda^2(T) \, (T_c - T)$ near T_c. This relation is illustrated by the dotted curve in figure 2, which extrapolates to zero at the same T_c as measured in low field. Thus, it is evident that the data are well behaved. The downward curvature at lower temperatures is expected. Fitting these data to the BCS temperature-dependence in the clean limit yields the solid curve shown in figure 2, with $\lambda(0) = 110$ nm. This dependence provides a better description than the BCS dirty limit, two fluid, or strong coupling dependencies. Due to increasing hysteresis of the sample, the data became less reliable for $T \leqslant 20$ K and were excluded from the fit. Assuming other temperature dependencies provides somewhat different extrapolated values, of course, as illustrated by the simple parabolic fit (dotted wave) in figure 2.

the sample, yielding the value $\langle d^4 \rangle^{\frac{1}{4}} = 12 \, \mu$m. The total volume of superconductor was obtained from its mass (6.6 mg) and the x-ray density of 2.7 g cm^{-3}. In addition to making the equilibrium signal more visible, powdering of the sample also insures that the critical currents flow in a better defined, intragranular geometry; this procedure removes the uncertainty about the size of effective current path that is inherent in studies of low density, porous materials.

The materials were studied magnetically in a SQUID-based magnetometer, at temperatures $T = 5$–300 K, in applied magnetic fields $\mu_0 H$ up to 6.5 T. The measurement system contains a sixth-order solenoid whose high homogeneity was independently verified; we also confirmed that field changes were monotonic in time, with no 'overshoot.' Scans of 3 cm were used. In the vortex state, we determined the volume magnetization $M(T, H, t)$ as function of H, T, and time t. (To convert M in Gauss into units of A m^{-1}, multiply by 10^3). The equilibrium magnetization M_{eq} was obtained by averaging the measurements in increasing and decreasing field, as illustrated below; in all cases, the background moment from the epoxy was removed by subtracting the temperature-independent magnetic moment measured at 50 K. The same hysteresis loops provide the supercurrent density $J = 15 \, \Delta M/r$, where ΔM is the hysteretic difference in M (in G) measured in increasing and decreasing fields and where $r = \langle d^4 \rangle^{\frac{1}{4}}/2$, (in cm). For studies of the time dependence of J, the temperature was first stabilized and the sample was taken into the critical state by setting a large negative field; then the field was increased to 0.25 T, where the magnetic moment was measured versus time for a period of ~ 1 h; following this, the field was increased for similar measurements at 0.5, 1 and 2 T. After increasing the field to 4 T, a short series of measurements were conducted at the same fields in the decreasing branch of the hysteresis loop, to obtain *in situ* values of the average (equilibrium) moment.
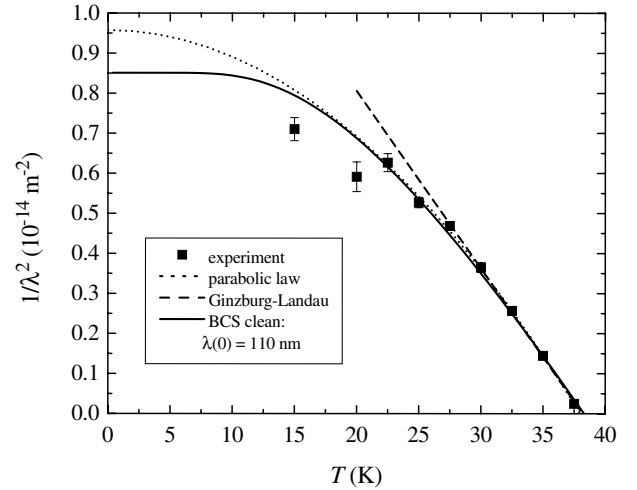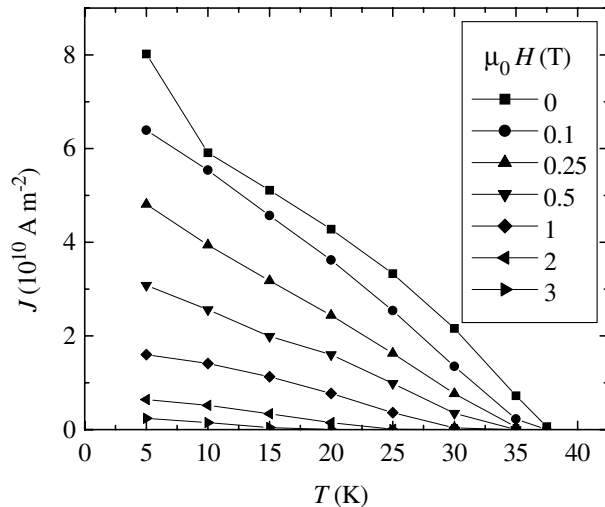
Figure 3. The temperature dependence of persistent current density J for MgB$_2$ in applied fields $\mu_0 H$, as shown.

Figure 4. The irreversibility line $B_{\mathrm{irr}}(T)$.

Figure 5. A plot of the normalized decay rate S versus temperature, for MgB$_2$ in applied fields of 0.25, 0.5, 1 and 2 T. Dotted vertical arrows show the irreversibility temperature for each case.

Next we consider the persistent current density J in the mixed state. Obtained from the magnetic hysteresis ΔM, results for J are shown in figure 3 as a function of temperature for several fixed fields. Several features are noteworthy. First, the temperature dependence is rather simple, with J falling off almost linearly as T increases. This contrasts sharply with the typical behaviour of J in high-T_c superconductors, which often exhibit a quasi-exponential fall-off associated with rapid thermally activated flux creep. A second feature is the relatively high scale of the current density, multiples of 10^{10} A m^{-2} at low temperatures. This is significantly higher than the values often reported for bulk ceramic materials, where porosity and possibly secondary phases may interfere with the intergrain transport of current. To put the present values into context, let us consider the depairing current density

$$J_{\mathrm{D}} = c\varphi_0 \left(12\sqrt{3}\pi^2\lambda^2\xi\right)^{-1} \approx 170 \times 10^{10}\mathrm{A\,m^{-2}},$$

here we use the above result for λ and the value $\xi = 5.2$ nm from Finnemore *et al* [7]. Relative to J_{D}, the experimental current densities in figure 3 are entirely reasonable, with J being approximately 5 % of the depairing current density. Thus, one may expect realized current densities to approach or exceed 10×10^{10} A m^{-2} with improved synthesis of thin films and bulk materials, both in transport and magnetic studies.

With increasing temperature and/or magnetic field, the current density decreases and eventually becomes immeasurable. Operationally, this defines an irreversibility line $B_{\mathrm{irr}}(T)$ which is shown in figure 4. These values were obtained from data like that in figure 3, using a criterion of 3×10^7 A m^{-2}, at which J has deteriorated by three orders of magnitude from its low temperature level. Again a simple, almost linear dependence is found, which is similar in magnitude and form to the $B_{\mathrm{irr}}(T)$ reported earlier [7].

Now we turn to the temporal stability of the persistent currents. A convenient measure is the (dimensionless) normalized decay rate $S = -\,\mathrm{d}\ln(J)/\mathrm{d}\ln(t)$, obtained from the time-dependent measurements. Experimentally, the decays we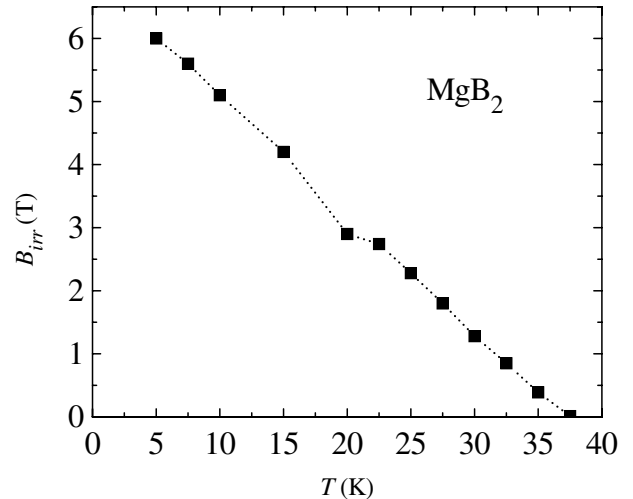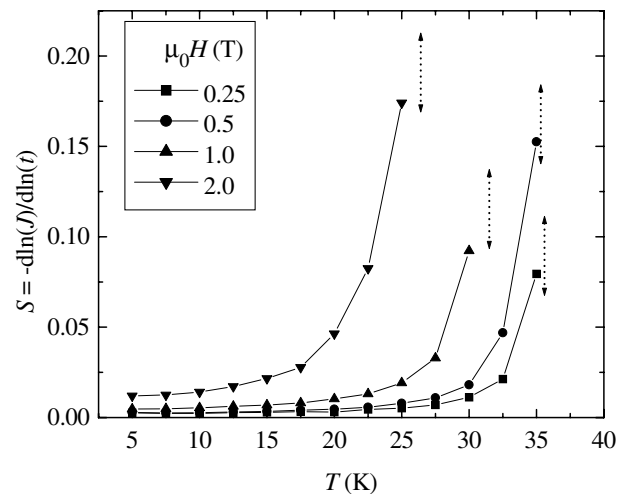re logarithmic in time and values for S were obtained from linear fits to plots of $\ln(J)$ versus $\ln(t)$. Estimates of the 'settling time' τ_0 needed to reach logarithmic decay [8] yielded values < 1 s, much below the measurement time scale. Figure 5 presents S versus T in fields of 0.25–2 T. The decay at low temperatures is quite small, with values near 0.003. In terms of a power-law relation between the electric field E and current density J with $E = E_0(J/J_0)^n$, the exponent n is simply related to S with $(n - 1) = 1/S$. This would imply that $n \sim 300$, corresponding to very steep and sharp curves in transport I–V studies. With increasing temperature, thermally activated depinning of vortices becomes more probable and S increases. Current decay typically accelerates as a system approaches the irreversibility line. The same feature is found here, as shown by the vertical arrows in figure 5 that mark respective irreversibility temperatures taken from figure 4. Just below these temperatures, S is increasing rapidly, while above them the decay is so rapid that J is unmeasurable on the time scale of the experiment. Bugoslavsky *et al* [9] have observed a similar rapid increase in flux creep as MgB$_2$ approaches the irreversibility line.
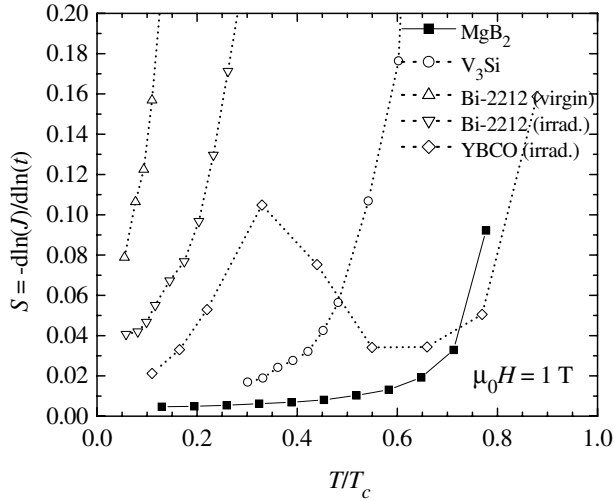
Figure 6. The normalized decay rate S versus reduced temperature T/T_c for several superconductors in an applied field of 1 T. The $Bi_2Sr_2CaCu_2O_x$ materials were c-axis textured tapes, either as-synthesized or irradiated with 0.8 GeV protons to create randomly oriented columnar defects with area density ('matching field') $B_\phi \sim 1.4$ T. The $YBa_2Cu_3O_7$ single crystal contains columnar defects with $B_\phi = 4.7$ T from 1 GeV Au ion irradiation. For the HTSC materials, the field was applied parallel to the c-axis.

It is useful to provide some context for the low decay rates observed in magnesium diboride. Thus we consider S as a function of reduced temperature T/T_c for some representative superconductors, all in an applied field of 1 T. The results are shown in figure 6. Materials include a V_3Si single crystal ($T_c = 16.6$ K), tapes of $Bi_2Sr_2CaCu_2O_x$ both as-synthesized and containing randomly oriented columnar defects created by fission fragments induced by 0.8 GeV protons [10], and a single crystal of $YBa_2Cu_3O_7$ containing parallel columnar defects formed by 1 GeV Au ions [11]. The striking feature is that the MgB_2 decay rate lies below all of these for nearly the entire range of reduced temperature. Compared with the HTSC materials, S at low temperatures is smaller by a factor of 3–10, and lies below that of the A15 phase crystal as well.

While the relative positions of the materials in figure 6 will differ in differing magnetic fields, it is encouraging and noteworthy that the persistent current density in MgB_2 is highly stable in magnetic fields of significant technological importance. In conclusion, studies of the reversible and irreversible magnetization of isolated MgB_2 particles show that the scale of current density in even as-synthesized materials is high, approximately 5% of the depairing current density. In magnetic fields of technological relevance, the supercurrents are highly stable in time over a significant temperature range.

Acknowledgments

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