Flux–creep activation energy for pure and SiC doped MgB$_2$ by ac-susceptibility measurements

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Abstract. ac susceptibility measurements were performed on pure and SiC-doped MgB$_2$ prepared by field assisted sintering technique in the temperature range between 5 and 50 K. The activation energy for flux–creep $U$ for both samples was determined at dc-fields of 0, 0.5, and 5 T, respectively, from the frequency dependence of the imaginary part of the ac-susceptibility $\chi''$ data measured in the range 17-9997 Hz. It was found that at 0 and 0.5 T, the activation energy of the SiC-doped sample is almost half the energy of the pristine sample whereas at 5 T the activation energies of the two samples become almost equal. A similar behaviour show the $H_p(T)$ lines as obtained from susceptibility data in the range 0-9 T. For fields lower than 7.5 T, the $H_p$ field of the doped sample is lower than for the undoped MgB$_2$. At $\sim$7.5 T, the $H_p(T)$ for SiC doped MgB$_2$ crosses over the $H_p(T)$ of pure MgB$_2$ and for higher fields the doped samples exhibits a higher irreversibility $H_p$. This field dependence was extracted from the evolution of the $\chi''(T)$ data. This behaviour is a consequence of the morphology of the sample with a large spread in grain size.

1. Introduction

The new MgB$_2$ superconductor is currently considered a reliable choice for large scale applications of superconductivity in helium-free refrigerated systems. It displays intermediate characteristics between low and high temperature superconductors. This new physics is due to its unique two-band superconductivity. This outstanding characteristic makes possible the control of the upper critical field $H_{c2}$ by the doping-triggered interplay of the interband/intraband scatterings [1,2]. Substitutions, incorporation of nano-particles or irradiation substantially also enhance pinning force. The fabrication methods are as well important for the final characteristics of the samples. The discovery of the MgB$_2$ superconductor [1] with a critical temperature of 39 K, started a thorough experimental and theoretical investigation of this material. For practical applications, an important parameter is the critical current density, which is ultimately related to the motion of vortices over the pinning centers (creep). Understanding creep process in MgB$_2$ [2] is the key toward the successful promotion of this material as a reliable alternative for the dissipation-free current transport. Simultaneously, intensive efforts have been expended to improve the critical current density by appropriate dopings [3-5].
A particular feature of MgB$_2$ is the doping induced increase of the critical current density at high magnetic field even though the doped material displays no enhancement or even a slight suppression of $J_c$ at low magnetic fields.

In this paper, we report the results of ac-susceptibility measurements of pristine and doped MgB$_2$ polycrystalline samples fabricated by field assisted sintering technique (FAST) in an attempt to find the origins of this remarkable property. We interpret the data as resulting from the large spread in the grain size. Our finding is supported by the microstructural data that have been already reported [6] in Si and SiC-doped samples.

2. Experimental

Polycrystalline samples of MgB$_2$ (MB) and SiC-doped MgB$_2$ (MBSC) were prepared from commercially available powders of MgB$_2$ (2.3 µm, Alpha Aesar), SiC (45 nm, Merck). In the case of doped samples, the molecular ratio between MgB$_2$ and dopant was 95/5. The samples were sintered using a “Dr Sinter” (Sumitomo Coal Mining Co, Japan) machine. The preparation details were published elsewhere [7]. All samples had identical sizes 7.7×3.3×0.8 mm$^3$ in order to avoid differences due to the samples geometry, and almost equal masses of 0.53 g. In this way, we were able to compare in a direct way the susceptibility data of different samples.

ac-susceptibility measurements were performed using an Oxford Instruments cryostat of the MagLab 2000 System with the ac field of amplitude $H_{ac} = 1$ Oe at frequencies between 17 Hz and 9997 Hz and dc-fields $H_{dc}$ in the range 0 – 9 T. Zero-field-cooling (ZFC) measurements were performed by first cooling the sample in zero magnetic field down to the starting temperature, then the external field was applied and the sample was warmed up. Both $H_{ac}$ and $H_{dc}$ were applied parallel to the longest side of the sample.

3. Results and discussion

Figure 1 shows the temperature $T$ dependence of the out-of-phase term of the fundamental harmonic $\chi''$ of the undoped sample (MB) in zero dc-field. The data were taken at different frequencies $f$, namely, at 17, 100, 1000 and 9997 Hz. This procedure allowed us to obtain the activation energy $U$ for the vortex creep using the frequency dependence of the peak temperature $T_p$ [8]:

$$f = f_0 \exp\left[ -\frac{U}{k_B T_p} \right] ,$$

where $f_0$ is a characteristic frequency and $k_B$ is the Boltzmann constant.

Using the Arrhenius plot, specifically, $1/T_p$ vs. $\ln f$ (see inset to Fig. 1) we obtained an (average) activation energy of 6.8 eV for the undoped MgB$_2$ and almost half of this value for the MBSC sample, i.e., 3.7 eV in zero applied dc-field.

As regarding the finite dc-fields, a reliable interpretation for weak fields $H_{dc} \leq H_{ac}$ is hard to complete. A considerable simplification is obtained for fields $H_{dc} \gg H_{ac}$, when the Bean’s model depicts the field penetration as occurring in symmetrical way. Thus, we can use the eq. (1) to obtain the field dependence of the activation energy from the ac-susceptibility data taken at different fields $H_{dc} \gg H_{ac}$. The $\chi''(T)$ data for the sample MBSC measured at $H_{dc} = 5$ T with an ac-field of amplitude of 1 Oe and four frequencies, namely, $f = 17, 100, 1000$ and 9997 Hz are shown in Figure 2. It is noticeable the strong frequency dependence of the $\chi''(T)$ curves.

The dependence of the activation energy on the applied field, temperature, and frequency, and current density is generally factorized as follows [9]:

$$U(T_p, H_{dc}, j) = U(T_p)U(H_{dc})U(j) = k_B T_p \ln\left[ \frac{1}{f_0} \right]$$

(2)
where \( t_0 \) is the time scale constant and \( j = H_{ac}/d \) (\( d \) is the sample size) is the critical current density. Therefore, the fit of \(-\ln(f)\) vs. \( U(T_p)/k_B T_p \) data should be a straight line with the slope \( U(j,H_{dc}) \):

\[
U(j,H_{dc}) = -\frac{d(\ln f)}{d(U(T_p)/k_B T_p)} .
\]

An explicit temperature dependence of the activation energy can be written as [2,10]:

\[
U(T) \propto \left[ 1 - \left( \frac{T}{T_{ons}} \right)^{\gamma} \right]^{-2},
\]

where \( T_{ons} \) is a characteristic temperature, which in the most natural way can be taken as the irreversibility temperature. \( T_{ons} \) for both sample was obtained from the onset temperature of the third harmonic \( |\chi_3(T)| \) (data not shown) measured at 1000 Hz and very small ac-fields (1 Oe). We obtained for the pristine MgB\(_2\) a \( T_{ons} \) of 37.02 K and 29 K at \( H_{dc} = 0.5 \) T and, respectively, 5 T and for the MBSC sample 35.39 K and 28.22 K for the same applied fields.

With the above data in mind, we can find the field dependence of the activation energy using the equations 5-7. Figure 3 shows the plot of \(-\ln(f)\) vs. \( U(T_p)/k_B T_p \) at both fields \( H_{dc} = 0.5 \) and 5 T and \( j \) (at \( H_{ac} = 1 \) G) = 10 A/cm\(^2\) for both samples. The linear fit provides the activation energies \( U(j,H_{dc}) \) at \( H_{dc} = 1 \) Oe, \( H_{dc} \). At small fields, these values are \( U(j = 10 \) A/cm\(^2\), \( H_{dc} = 0.5 \) T) = 12.2 eV for the MB sample and 6 eV for the doped MBSC sample, respectively. In high field regime we obtained \( U(j = 10 \) A/cm\(^2\), \( H_{dc} = 5 \) T) = 0.82 eV for the pristine MgB\(_2\) and 0.85 eV for MBSC sample, respectively. A clear tendency toward the reversal of the activation energies for flux creep occurs at high fields, which is conspicuous in Fig. 3.

A similar evolutions is noticeable in the \( H_d(T) \) curves for MB and MBSC samples as obtained from the peak position in of the \( \chi_1''(T) \) data taken at different values of \( H_{dc} \) (fig. 4) where it is a clear cross of the two curves at \( H = 7.5 \) T and approximately 23 K.

A hint of the reason of this inversion is given by a careful examination of the susceptibility data. We have plotted in Fig. 5 the data for both samples at different fields to illustrate the changes in which occur in susceptibility, mainly the out-of-phase contribution.
It is clear the different dc-field dependence of the two samples that is reflected in a continuous increase in amplitude of the response of MBSC sample.

We remind here that the imaginary part of the complex susceptibility is directly related to the ac-loss, which for a cycle is given by:

$$Q = \oint H_{ac} dM$$

With $M'' = V \chi'' H_{ac}^{\max}$ one obtains:

$$\chi'' \propto \frac{Q}{V(H_{ac}^{\max})^2}$$

The maximum $\chi''$ calculated with the critical state model is almost five times smaller than the maximum value of $\chi'$, which is $1/4\pi$. Experimentally, this is well confirmed in ceramics, where the transition in $\chi'$ is steep and displays an almost perfect diamagnetism. The smaller values of $\chi''$ are mainly connected to the low density of the samples and the smaller weight of the superconducting phase within sample. The width of the $\chi''$ curve is a good hint on transition length, hence, on the
degree of homogeneity of the sample. A sample with a large spread in grain size, at a given
temperature, displays only the signal coming from the grains larger than the field penetration length at
that temperature $\lambda(T)$. We also remind here that the position $T_p$ of the peak of $\chi''$ is used to obtain the
critical current density $J_c$. Thus, the behaviour of $\chi''$ as a function of $H$ and $T$ is the convolution of the size
grain, their density and their pinning capability. Due to the longer coherence length, the
granularity is less important in good MgB$_2$ samples than in cuprates, where the grain borders are less
transparent to the supercurrent. However, the granularity effects cannot be avoided in all samples.

First, we scrutinize the case of high temperature and small fields. Figure 5 shows that the sample
MB has the characteristics of an almost ideal superconductor in zero applied field. Specifically, it has a
narrow transition with the ratio $\chi_1'/\chi_1'' = 4.6$ close to the theoretical value of 5. The small width and
the relative high amplitude of $\chi_1''$ advocate for a narrow distribution of both grain size and pinning
energy, i.e., the field penetrates all grains at almost the same temperature. This is also the reason of the
slow field dependence of the peak amplitude at fields $H_{dc} >> H_{ac}$.

The susceptibility curves of the MBSC sample show a broad width and small amplitude in an
agreement with a large spread in the grain size and in their pinning energies. When the temperature
increases, the field penetrates first the smallest grains and those with the weakest pinning. Additionally,
the progressive increase of the amplitude $\chi''$ with the increase of $H_{dc}$ suggests the
generation of new pinning centers, most likely induced by field in the weakest superconducting grains.

At very high fields (small $T$), both samples display similar homogeneity as mirrored by their
almost equal transition width. At $H = 9$ T the amplitudes of $\chi_1''$ for the two samples almost even, and
the peak position even changes the order (MBSC peak occurs at a higher temperature than MB peak).
This pinning enhancement can occur on the account of weak superconducting grains, in which the
field suppresses the superconductivity and transforms them in good pinning centers. Additionally, the
internal defects of grains (dislocation networks) become less transparent to superelectrons in high
fields, but also very strong pinning centers.

The above assumption on the role of grain dispersion is in good agreement with the structural
investigation on the same samples [6] that showed a better homogeneity in the grain size in the case of
MgB$_2$ sample as compared with SiC-doped sample. The latter, in addition to large size dispersion,
shows also a poor connectivity of the grains. Also in Si and SiC-doped MgB$_2$, transmission electron
microscopy data reported a plethora of defects: inclusions, MgO and Mg$_2$Si nanoparticles, nanodomain
structures [3] as well as intragrain dislocations and dispersed nanosized impurities [5].

4. Conclusions

Using the frequency dependence of the $\chi_1''(T)$ we find that both in fields up to 0.5 T the activation
energy of flux creep for pristine MgB$_2$ polycrystalline sample is higher than that of the SiC-doped
sample, whereas at high fields ($H_{dc} = 5$ T) the creep activation energy for both samples are almost
equal. The dependence $H_p(T)$ follows the same evolution. A cross of the $H_p(T)$ plots for the two
samples occurs at 7.5 T and low temperatures (23 K) when the activation energy for the SiC-doped
samples becomes higher than for the pristine MgB$_2$. The shape and position of the $\chi_1''(T)$ curves
suggested that the high spread of the grain size distribution as well as their spatial distribution in the
doped sample is the cause of the pinning depression at high temperature and small field but also is
responsible for the pinning enhancement at low temperature and high fields.

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