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## FLUX DYNAMICS OF MgB<sub>2</sub> SUPERCONDUCTOR BY AC SUSCEPTIBILITY MEASUREMENTS

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### ABSTRACT

Flux dynamics of an MgB<sub>2</sub> bulk sample have been studied by systematic measurements of the fundamental and third harmonic ac susceptibility. The irreversibility line  $T_{irr}(B)$  has been obtained by fitting the onset temperature  $T_{on}(f, B)$  of the third harmonics at different frequencies to  $T_{on}(f, B) = C(2\pi f)^{1/[(z-1)\nu]} + T_{irr}(B)$ . The flux creep activation energy  $U(j, B, T)$  has been derived as  $U_0[1 - (T/T_{irr})^2]^2(B/B_0)^{-n}(j/j_0)^{-\mu}$ , with  $n=1.33$  and  $\mu=0.2$ . The steep decline of the activation energy as a function of the magnetic field  $U(B) \propto B^{-1.33}$  leads to the steep drop in the critical current density with magnetic field. The relaxation of the current density  $j(t)$  of the sample has been detected in a time window of  $10^{-4} \sim 10^{-1}$  s, as the frequency is in the range of 10 Hz-10 kHz, which extends the time window  $1 \sim 10^4$  s of the conventional dc magnetic relaxation to a much smaller one. The obtained activation energy and the irreversibility line indicate that the pinning properties of the MgB<sub>2</sub> sample will need to be improved for practical applications.

### INTRODUCTION

The new superconductor MgB<sub>2</sub> [1] with its transition temperature at 39 K has been the subject of intensive study since its discovery [2-21]. The critical current density, one of the most important parameters in considering superconductors for practical applications, has been determined to be about  $10^6$  A/cm<sup>2</sup> at 0 T and 4.2 K in bulk samples [6] and  $8 \times 10^6$  A/cm<sup>2</sup> at 0 T and 5 K in thin films [22]. Although these zero field values are quite exciting compared to the high temperature superconductors, they are found to decrease rapidly with the applied magnetic field [2-9]. The critical current density is determined by the pinning properties of the sample as well as by the flux motion, because the motion of the vortices over pinning centres (flux creep) in the superconductor induces dissipation and reduces the critical current density  $j_c$ . It is the flux creep that sets the limiting critical current density

in superconductors. It is thus essential to study the activation energy against flux motion, in order to understand the underlying mechanism resulting in the rapid decreasing relationship between the critical current density and magnetic field, and therefore to enhance the current carrying capacity of this new material.

On the other hand, the irreversibility line can be obtained by several methods. However, as has been pointed out by Deak et al. [23], not all the methods are reliable. For the  $\text{MgB}_2$  samples, the irreversibility line was usually determined by means of magnetization measurement [2,6], i.e., the close point of the magnetization hysteresis loop at high magnetic field. However, the irreversibility line determined using this method depends on the sweep rate of the applied magnetic field  $dB/dt$ , larger  $dB/dt$  results in higher irreversibility field. This is similar to the ac susceptibility measurements, where the results depend on the frequency of the applied ac magnetic field. A more reliable method using ac susceptibility to determine the irreversibility line has been proposed by Deak et al. [23]. The key point is to measure the third harmonic ac susceptibility at different frequencies, and then the irreversibility temperature  $T_{irr}$  can be obtained by fitting the onset temperature  $T_{on}(f, B)$  of the third harmonic ac susceptibility to

$$T_{on}(f, B) = C(2\pi f)^{1/[(z-1)\nu]} + T_{irr}(B) \quad (1)$$

where  $C$  is a constant,  $\nu$  and  $z$  are vortex-glass scaling parameters, and  $(z-1)\nu$  is used as a fitting parameter. The obtained irreversibility line is then frequency independent.

In this paper, we investigate the flux creep activation energy in  $\text{MgB}_2$ , and determine its dependence on the current density, the magnetic field, and the temperature by measuring the real  $\chi'(T)$  and imaginary  $\chi''(T)$  parts of the ac susceptibility at different ac field amplitudes, frequencies and dc magnetic fields. The irreversibility line is obtained using the third harmonic ac susceptibility technique.

## EXPERIMENTAL

All measurements have been performed on a  $\text{MgB}_2$  bulk sample ( $T_c = 38.6$  K,  $\Delta T_c < 1$  K by ac susceptibility in an ac field of 1 G and frequency 117 Hz.), which was prepared by conventional solid state reaction [24]. High purity Mg and B (amorphous) with a nominal composition ratio of Mg:B=1.2:2 were mixed and finely ground, then pressed into pellets 10 mm in diameter with 1-2 mm thickness. Extra Mg was added in order to make up for loss of Mg at high temperatures. These pellets were placed on an iron plate and covered with iron foil, then put into a tube furnace. The samples were sintered at temperatures between 700 and 1000°C for 1-14h. A high purity Ar gas flow was maintained throughout the sintering process. A sample with dimension of  $2.18 \times 2.76 \times 1.88$  mm<sup>3</sup> was cut from the pellet. Phase purity was determined by XRD and grain size by SEM. Only a small amount of MgO (less than 10%) was found and the grain size was determined to be about 200  $\mu\text{m}$ .

The ac susceptibility measurements were carried out using a Quantum Design PPMS.

## RESULTS AND DISCUSSIONS

Shown in FIG 1a are typical  $\chi'(T)$  and  $\chi''(T)$  curves for the  $\text{MgB}_2$  bulk sample at  $B_{dc} = 1$  T,  $f = 1117$  Hz and different ac field amplitudes  $B_{ac}$  indicated. As  $B_{ac}$  is increased

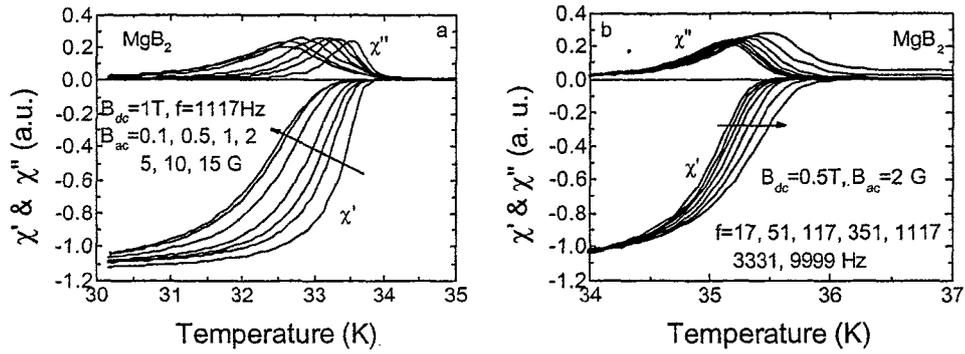


FIGURE 1. a)  $\chi'(T)$  and  $\chi''(T)$  curves of the  $\text{MgB}_2$  sample at  $B_{dc}=1$  T,  $f=1117$  Hz and  $B_{ac}=0.1, 0.5, 1, 2, 5, 10, 15$  G (from right to left). b)  $\chi'(T)$  and  $\chi''(T)$  curves of the sample at  $B_{dc}=0.5$  T,  $B_{ac}=2$  G and  $f=17, 51, 117, 351, 1117, 3331, 9999$  Hz (from left to right).

the transition shifts to lower temperatures with increased width. In FIG 1b, we show the effects of the frequency on the ac susceptibility of this  $\text{MgB}_2$  sample. Contrary to the effects of  $B_{ac}$ , as  $f$  is increased, the transition shifts to higher temperatures and the transition width broadens. All the characteristics shown in FIG 1 for the  $\text{MgB}_2$  sample are similar to what have been observed in high temperature superconductors [25,26] and predicted from theoretical calculations [27]. This is understandable, because ac susceptibilities at different dc magnetic fields, ac field amplitudes and frequencies reflect a common phenomenon, i.e., flux dynamics in type-II superconductors.

A measurement of the superconducting transition by means of the ac susceptibility  $\chi = \chi' + i\chi''$  typically shows, just below the critical temperature  $T_c$ , a sharp decrease in the real part of the susceptibility  $\chi'$ , a consequence of diamagnetic shielding, and a peak in the imaginary part of the susceptibility  $\chi''$ , representing losses. The peak in  $\chi''$  will occur when the flux front reaches the centre of the sample. It follows that the position of the peak in  $\chi''$  will also strongly depend on temperature, dc field, ac field amplitude and frequency. The criterion for the peak in  $\chi''$  is [28]

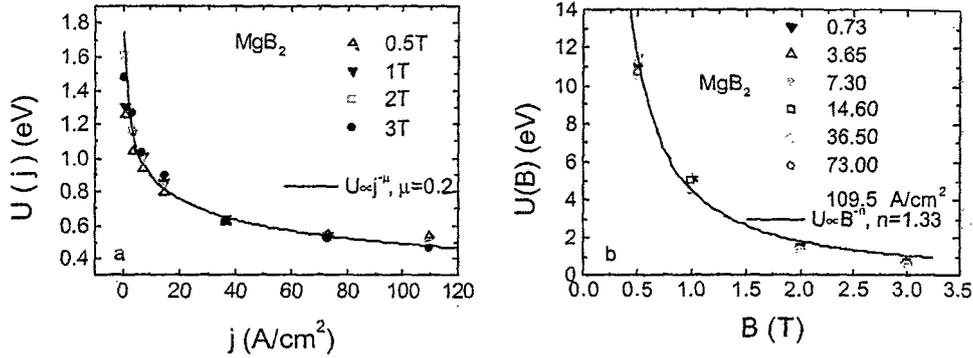
$$U(T_p, B_{dc}, j) = k_B T_p \ln \left( \frac{1}{f_{peak} t_0} \right) \quad (2)$$

where the time scale  $t_0 = 4\pi\mu_0 H_{ac}^2 / \rho_0 j^2(f)$  [28],  $\rho_0$  is the prefactor in the Arrhenius law  $\rho = \rho_0 \exp[-U(j)/k_B T]$ ,  $T_p$  is the peak temperature in the  $\chi''$  curve and  $k_B$  the Boltzmann constant.

It has been shown [27] by numerical calculation that during the penetration by the ac magnetic field into a superconductor, the magnetic field profile can be regarded as a straight line, so at the peak temperature the current density can be approximated as

$$j = \frac{H_{ac}}{d} \quad (3)$$

where  $d$  is the sample size.



**FIGURE 2.** a) Activation energy  $U(j) \propto U(j, B) \times B^{1.3}$  as a function of the current density for the  $\text{MgB}_2$  sample at various dc magnetic fields. Solid line is the fitting curve  $U(j) \propto j^{-0.2}$ . b) Activation energy  $U(B) \propto U(j, B) \times j^{0.21}$  as a function of the magnetic field for the  $\text{MgB}_2$  sample at various current densities. Solid line is the fitting curve  $U(B) \propto B^{-1.33}$ .

A plot of  $-\ln(f_{peak})$  versus  $U(T_p)/k_B T_p$  should thus be a straight line with the slope of  $U(j, B_{dc})$ , since

$$\frac{U(j, B_{dc}, T)}{k_B T} = \frac{U(T)}{k_B T} U(j, B_{dc}) = -\ln(f) - \ln(t_0). \quad (4)$$

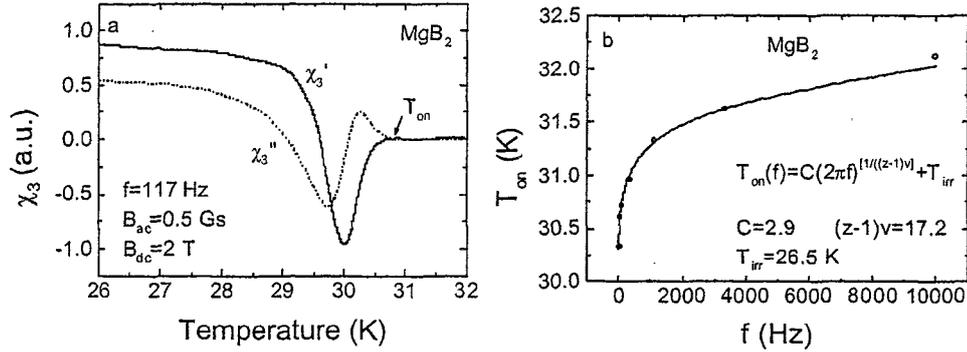
By varying the ac amplitude and then using equation (3) to determine the current density, one can then reconstruct the current density dependence of the activation energy  $U(j)$ . Using the ac method the usual difficulty in conventional relaxation measurements of having only a very limited time window ( $1 \sim 10^4$  s) can be overcome by extending the latter to smaller values of  $10^{-5} \sim 10^{-3}$  s ( $f = 100$  kHz  $\sim$  1 kHz) [25,26].

In order to account for the explicit temperature dependence of the activation energy, we choose a form of temperature scaling function

$$U(T) = [1 - (T/T_{irr})^2]^2 \quad (5)$$

where  $T_{irr} = 35.4, 32.3, 26.5, 20.2$  K for  $B_{dc} = 0.5, 1, 2, 3$  T respectively is taken from the irreversibility line (FIG 4).  $U(T)$  changes slightly with temperature for  $T \ll T_{irr}$  and drops rapidly as  $T$  approaches  $T_{irr}$ . A detailed discussion on choosing the function  $U(T)$  has been given by McHenry et al. [29].

FIG 2a shows the activation energy  $U(j) \propto U(j, B) \times B^{1.3}$  as a function of the current density for the  $\text{MgB}_2$  sample at various dc magnetic fields. As can be seen from FIG 2a, we have obtained a universal curve  $U(j)$  by scaling the data by  $B^{1.3}$ . The slight scattering at low current density may result from the field dependent critical current density  $j_c(B)$ . Note that  $B_{ac}$  has been changed to  $j$  using equation (3), where  $d$  is the sample size rather



**FIGURE 3.** a)  $\chi_3'(T)$  and  $\chi_3''(T)$  curves of the  $\text{MgB}_2$  sample at  $B_{dc}=2$  T,  $f=117$  Hz and  $B_{ac}=0.5$  G. b) The onset temperature of the third harmonics as a function of the frequency at  $B_{dc}=2$  T and  $B_{ac}=0.5$  G (symbols), solid line is a fit to equation (1).

than the grain size, because it has been reported [2] that current flow in  $\text{MgB}_2$  is strongly linked. The current density  $j$  obtained here is also very close to what has been derived using magnetization measurements [24].

From the best fit of the data in FIG 2a, we derived the current density dependent activation energy  $U(j) \propto j^{-0.2}$ , which is highly non-linear. This result suggests that the  $I-V$  curve of  $\text{MgB}_2$  should also be highly non-linear, because using the Arrhenius rate equation, we have  $E = Bv = Bv_0 \exp[-U(j)/k_B T] \propto \exp(-j^{-\mu})$ . Non-linear  $I-V$  characteristics have been experimentally observed in  $\text{MgB}_2$  [12]. On the other hand, the relaxation of the current density or the magnetization can be derived from equation (2) as  $j(t) \propto [\ln(t/t_0)]^{-1/\mu}$ , which is also a non-linear function of  $\ln(t/t_0)$ .

As can be seen from equation (4), with the current density  $j$  fixed, we can also derive the activation energy as a function of the dc magnetic field  $U(B)$ . The results are summarized in FIG 2b, where the activation energy  $U(B) \propto U(j, B_{dc}) \times j^{0.2}$  is plotted as a function of the magnetic field for the  $\text{MgB}_2$  sample at various current densities. As can be seen from FIG 2b, we have also obtained a universal curve by scaling the data by  $j^{0.21}$ . This current density dependence is consistent with the one derived in FIG 2A. As the scaling factor  $B_0$  [equation (6)] for  $B$  is current density independent, we can see that the scaling of  $U(B)$  is much better than that of  $U(j)$  shown in FIG 2a. The solid line in FIG 2b is a fit to the power law  $U(B) \propto B^{-1.33}$ . The obtained  $U(B)$  is also consistent with the one derived from scaling in FIG 2a. The self-consistent scaling of  $U(j, B)$  shown in FIG 2a and FIG 2b suggests that the separation of the activation energy  $U(j, B, T)$  to  $U(j)U(B)U(T)$  is quite reasonable. The final expression for the temperature, field and current density dependent activation energy is given by

$$U(T, B, j) = U_0 \left[ 1 - \left( \frac{T}{T_{irr}} \right)^2 \right]^2 \left( \frac{B}{B_0} \right)^{-n} \left( \frac{j_0}{j} \right)^\mu \quad (6)$$

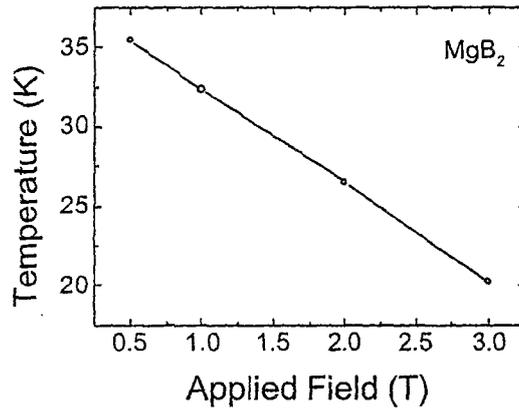


FIGURE 4 The irreversibility line of the MgB<sub>2</sub> sample, solid line is just guide to the eye.

where  $U_0$ ,  $B_0$  and  $j_0$  are scaling values, and the exponents  $n$  and  $\mu$  are determined to be 1.33 and 0.2 respectively.

As for the magnetic field dependence of the activation energy, a  $B^{-1}$  dependence has been previously derived using the Anderson-Kim model of the activation energy combined with the Ginzburg-Landau expressions for the coherence length, thermodynamic critical field and depairing critical current density, etc. [30,31]. Such a  $B^{-1}$  dependence has been observed in a La<sub>1.86</sub>Sr<sub>0.14</sub>CuO<sub>4</sub> single crystal with weak pinning centres by McHenry et al. [29]. And for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> samples with strong pinning centres, such as twin planes, stacking faults or Y<sub>2</sub>BaCuO<sub>5</sub> inclusions, a  $U(B) \propto B^{-0.5}$  has been derived by both ac susceptibility [26] and dc magnetization measurements [32,33].

For the new superconductor MgB<sub>2</sub> on the other hand, we find a  $U(B) \propto B^{-1.33}$  dependence showing that the activation energy decreases even faster with increasing magnetic field, compared to weakly pinning high temperature superconducting La<sub>1.86</sub>Sr<sub>0.14</sub>CuO<sub>4</sub> single crystal. The weakening of the activation energy with increasing magnetic field may be the reason why the critical current density drops steeply as the magnetic field increases, as has been observed by dc magnetization measurements [2-6,9].

Shown in FIG 3a are  $\chi_3'(T)$  and  $\chi_3''(T)$  curves of the MgB<sub>2</sub> sample at  $B_{dc} = 2$  T,  $B_{ac} = 0.5$  G and  $f = 117$  Hz. The onset temperature is marked on the figure. The  $\chi_3$  curves are different from what have been observed in strong pinning high temperature superconductors, such as YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub>. This topic will be discussed in one of our forthcoming works. Similar  $\chi_3$  curves at different magnetic fields, ac field amplitudes and frequencies have also been measured, the results are not shown here for simplicity. FIG 3b shows the measured onset temperature  $T_{on}$  versus the magnetic field (symbols), solid line is the best fitting using equation (1). As can be seen from the figure, equation (a) can fit the experimental data very well. Extrapolating the curve to  $f = 0$ , we can obtain the irreversibility temperature at  $B_{dc} = 2$  T. The irreversibility temperatures at  $B_{dc} = 0.5$ , 1 and 3 T have also been derived, which are shown together in FIG 4. Compared to high temperature superconductors, the irreversibility line of the MgB<sub>2</sub> sample is relatively low.

## SUMMARY

In summary, taking into account the relatively low irreversibility line and the weakening of the activation energy with increasing magnetic field, the pinning properties of MgB<sub>2</sub> will need to be enhanced for practical applications.

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