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Magnetic relaxation and critical current density of MgB_2 thin films

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The magnetic relaxation and critical current density have been measured on a MgB_2 thin film in a wide region of temperature with magnetic field up to 8 T. The irreversibility line has also been determined. It is found that the relaxation rate has a very weak temperature dependence below $1/2T_c$, showing a clear residual relaxation rate at 0 K, which cannot be easily explained as due to thermally activated flux creep. Furthermore, the relaxation rate has a strong field dependence. The flux dynamics of thin films is very similar to that of high-pressure synthesized bulks although the relaxation rate in thin film is systematically higher than that of a bulk sample. All the results here together with those from bulk samples suggest that the flux dynamics may be dominated by quantum effects, such as quantum fluctuation and tunneling.

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I. INTRODUCTION

The recently discovered new superconductor MgB_2 has become very attractive due to its potential applications.[1–4] One important issue, however, is concerned with which region on the field-temperature (H-T) phase diagram it can carry a large critical current density (j_c) and thus can be used in the future for industry. This j_c is controlled by the mobility of the magnetic vortices, and vanishes above the melting line between the vortex solid and liquid. This melting can be induced by a strong fluctuation of the vortex position by either the thermal effect or quantum effect. At $T=0$ K only the quantum fluctuation is left. A finite linear resistivity $\rho_{lin}=(E/j)_{j\rightarrow 0}$ will appear above this melting point, showing reversible flux motion. Most of the published results on the flux dynamics of the MgB_2 system were obtained from bulk samples.[5–9] In this paper we present an investigation of the flux dynamics on an MgB_2 thin film by the dynamical magnetic relaxation method.

II. EXPERIMENT

The thin films of MgB_2 were fabricated on (1102) Al_2O_3 substrates by using the pulsed laser deposition technique.[10] The amorphous B thin film was first deposited and then it was sintered at a high temperature in Mg vapor. The films are typically 400 nm thick with predominant c-axis orientation (the c axis is perpendicular to the film surface). A rectangular sample of size 2.1 mm$\times$4.9 mm was chosen for the magnetic sweeping measurement. The magnetic measurements were carried out by a Quantum Design superconducting quantum interference device (SQUID, model MPMS 5.5 T) and a vibrating sample magnetometer (VSM model 8T, Oxford 3001) at temperatures ranging from 2 K to T_c and external field up to 8 T. For the magnetic sweeping measurement the $M(H)$ curve was measured with different field sweeping rates (0.005 T/s–0.02 T/s) and integral time of 60–960 ms. The pressure of He gas in the sample chamber for thermal exchange was kept at 0.4 bar during the measurement. The superconducting transition is sharp with a transition temperature T_c of about 38 K as observed from the temperature dependence of the magnetization.

III. RESULTS

In Fig. 1 we show the magnetization hysteresis loops (MHL's) measured at temperatures ranging from 2 K to 37 K. The symmetric MHL's observed at temperatures up to 37 K indicate the dominance of the bulk current instead of the surface shielding current. The MHL's measured at low temperatures, such as from 2 K to about 10 K, show dense and small flux jumps in the low-field region, leading to a smearing of the superconducting critical current density. It is this

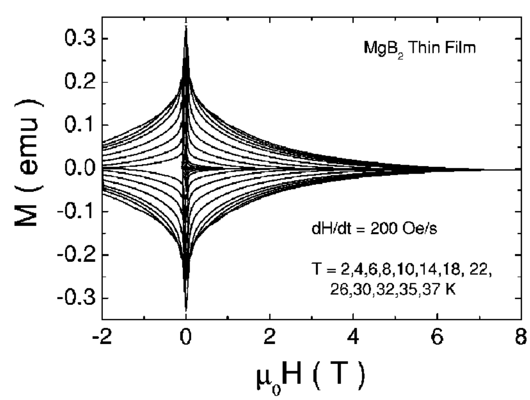

FIG. 1. Magnetization hysteresis loops measured at 2, 4, 6, 8, 10, 14, 18, 22, 26, 30, 32, 35, 37, and 38 K (from outer to inner). All curves here show a symmetric behavior indicating the importance of bulk current instead of surface shielding current. The MHLs measured at low temperatures (e.g., 2–10 K) are too close to be distinguishable. Dense and small flux jumps have been observed below 10 K near the central peak.

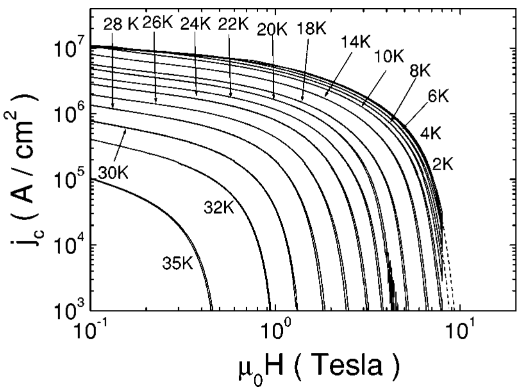

FIG. 2. Critical current density j_c calculated based on the Bean critical-state model. At each temperature the data have been measured with two field sweeping rates 0.02 and 0.01 T/s. The faster sweeping rate corresponds to a higher dissipation and thus higher current density. From these data one can calculate the dynamical magnetic relaxation rate Q. The $j_c(H)$ curves measured at low temperatures are very close to each other showing a rather stable value of H_{irr} when T approaches 0 K. We use a criterion of $j_c = 1000$ A/cm^2 to determine the irreversibility line.

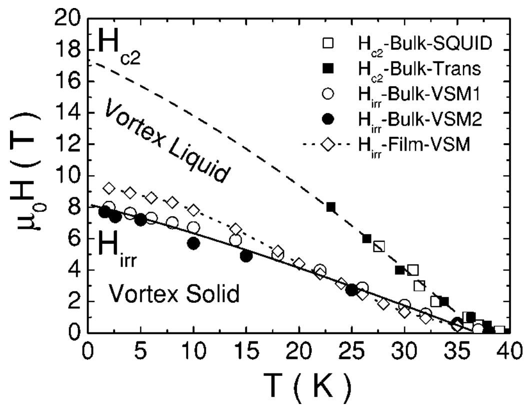

FIG. 3. H-T phase diagram for the superconductor MgB_2. The circles represent the bulk irreversibility lines H_{irr} of bulk samples: open and solid circles represent two measurements by the VSM on two bulk samples. The open diamond symbols represent the irreversibility line of the thin film. The squares represent the upper critical field $H_{c2}(T)$: solid squares are from resistive measurements; open squares are from the $M(T)$ measurements by the SQUID. All lines are a guide to the eye.

effect that makes the width of the magnetization ΔM below 10 K be anomalously smaller than that at 14 K, which is discussed elsewhere.[11] From these MHL's one can calculate j_c via $j_c = 20\Delta M/Va(1 - a/3b)$ based on the Bean critical-state model, where V, a, and b are the volume, width, and length of the sample, respectively. The result of j_c is shown in Fig. 2. It is clear that the magnetic critical current density j_c of our sample is rather high. For example, at $T = 18$ K and $\mu_0 H = 1$ T, we have $j_c = 2\times10^6$ A / cm^2, which is about one order of magnitude higher than that of high-pressure synthesized bulk samples.[6,7,12]

For investigating the flux dynamics the $j_c(H)$ curves have been measured with two different field sweeping rates of 0.02 and 0.01 T/s. It is interesting to note that although for bulk samples[4,7] there is a small tail on each MHL in the high-field region, here on the thin-film sample there is no such tail. Accordingly the $j_c(H)$ curves do not show a small tail in the high-field region. This further corroborates our earlier suggestion that the small tail observed in bulk samples is due to some secondary effect, such as some local regions with very strong pinning or the surface pinning by tiny grains. In the present thin-film sample with much better uniformity this effect has certainly disappeared. From the contour of j_c vs H shown in Fig. 2 one can see that all curves have two different regions. In the low-field region j_c drops slowly with H. When the field is increased further and exceeds a threshold j_c decreases drastically, showing a gradual setting in of the reversible flux motion. One can determine the phase transition line which separates the irreversible and the reversible flux motion by taking a criterion, here, for example, $j_c = 1000$ A / cm^2. The same criterion was used by Bugoslavsky *et al.* for bulk samples.[2] Since our measurement was done at a maximum field of 8 T, for low temperatures we use a reasonable method of extrapolation, i.e., to follow the tendency of j_c vs H at a higher temperature, for example, at 10 or 14 K, down to the criterion $j_c = 1000$ A / cm^2 to derive the irreversibility field. This method, although with some uncertainties for the values of $H_{irr}(T)$ at low temperatures, will lead us to derive an almost complete curve of $H_{irr}(T)$. The error bar of $H_{irr}(T)$ at 2 K is about ± 0.5 T. The irreversibility lines by following this method are shown in Fig. 3 together with that from a high-pressure synthesized bulk sample. Similar to what is found in bulk samples,[3,2,4,6,7] it is easy to see in the present thin film that $H_{irr}(T)$ extrapolates to a rather low field, for example, $H_{irr}(0) \approx 9.2 \pm 0.5$ T, while $H_{c2}(T)$ extrapolates to a much higher value (≈ 15 T) (Ref. 13) at zero K. There is a large separation between the two fields $H_{c2}(0)$ and $H_{irr}(0)$. This effect, observed in both bulk and thin-film samples, may suggest that the relatively low $H_{irr}(T)$ in MgB_2 is not due to the easy flux motion through some weak pinning channels; rather, it reflects probably a more intrinsic property, especially in a rather clean system.

IV. DISCUSSION

A. Large separation between $H_{irr}(0)$ and $H_{c2}(0)$ and possible evidence for a quantum vortex liquid in MgB_2

Following the hypothesis of a vortex liquid above $H_{irr}(T)$, we would conclude that there is a large region for the existence of a quantum vortex liquid at 0 K. This can be attributed to a quantum fluctuation effect of vortices in bulk MgB_2. Although the lowest temperature in our present experiment is 2 K, however, from the experimental data one cannot find any tendency for $H_{irr}(T)$ to turn upward to meet the $H_{c2}(0)$ at 0 K. This may indicate that the vortex melting in our present film is due to the strong quantum fluctuation which smears the perfect vortex lattice, leading to the vanishing of the shear module C_{66} of the vortex matter (probably within grains). Dense disorders will strengthen the shear module and thus enhance the irreversibility line; that is why the H_{irr} in the present thin film it is higher than that in the bulk samples (shown in Fig. 3). That the irradiation of protons by Bugoslavsky *et al.*[14] did not suppress but strongly increase j_c at a high field would suggest that the low value of $H_{irr}(T)$ measured in unirradiated bulk samples and the present thin film is not due to the weak links since otherwise the j_c value would drop even faster with increasing the magnetic field after the irradiation.

In order to investigate the flux dynamics in the vortex solid state below H_{irr} we have carried out a dynamical magnetic relaxation measurement. Assuming a uniform current density over the cross section of a superconducting ring, one can determine the superconducting current density j_c from the magnetic moment M via

$$M=\frac{1}{3}\pi j_c d(R_o^3-R_i^3), \quad (1)$$

where d is the film thickness, and R_o and R_i are the outer and inner radii of the ring, respectively. It is important to note that the magnetic moment M in Eq. (1) is understood as being due to only the superconducting current, excluding any additional contribution from a equilibrium magnetization. In other words, M is obtained by subtracting from the zero-field-cooled (ZFC) magnetic moment with the equilibrium magnetic moment as determined from the field-cooled (FC) process, i.e., $M=M$ (measured) $-M$ (equilibrium). For a superconducting thin film the equilibrium magnetic moment is normally negligible since the volume of the superconducting material is very small. Based on different external conditions, there are two techniques to measure the magnetic relaxation, namely, the so-called conventional and dynamical relaxation. The so-called conventional relaxation measures the time dependence of the superconducting current density j_c at a certain temperature and field.[15–17] After a waiting time when the magnetic field is fixed (or, say, field sweeping is stopped), the first data point is taken. For a conventional relaxation measurement, the total observation time should be very long. The second method is the so-called dynamical relaxation, i.e., to measure the MHL's with different field sweeping rates. One can easily understand the difference between these two methods from the electromagnetic response of a superconducting ring. The electromotive force in a ring is

$$E2\pi R=\pi R^2\frac{d(\mu_0 H)}{dt}-wdL\frac{dj_c}{dt}, \quad (2)$$

where E is the electric field established within the ring, $R=(R_o+R_i)/2$, H is the external field, $L=\mu_0 R[\ln(8R/w)-1/2]$ is the self-inductance of the ring,[18] and $w(=R_o-R_i)$ is the width of the ring. For the relaxation process, since $dH/dt=0$, the electric field can be determined by

$$E=-\frac{\mu_0 wd}{2\pi}\left[\ln\left(\frac{8R}{w}\right)-\frac{1}{2}\right]\frac{dj_c}{dt}. \quad (3)$$

This method is inapplicable when the irreversible magnetic signal is comparable to the equilibrium magnetization. For MgB_2 with a very narrow MHL this method shows a clear drawback. However, one can choose the so-called dynamical relaxation method.[19,20] This technique can be accomplished by using a sensitive VSM or a torque magnetometer. In the field sweeping process, if the field sweeping rate is high enough, the last term in Eq. (2) can be neglected; therefore, the electric field E can be determined by

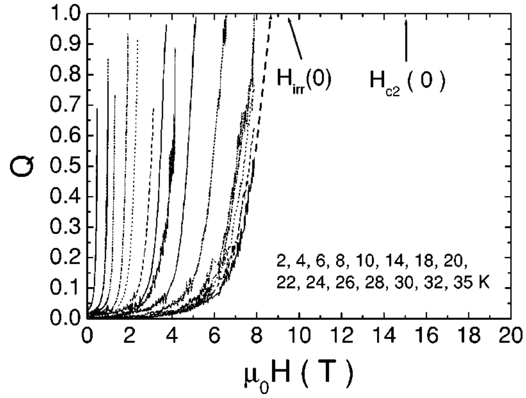

FIG. 4. Field dependence of the relaxation rate at temperatures of 2, 4, 6, 8, 10, 14, 18, 20, 22, 24, 26, 28, 30, 32, and 35 K. The dashed line is a guide to the eye for 2 K. It is clear that Q will rise to 100% at about 8.7 T at 2 K. Since H_{irr} is rather stable at low temperatures, it is safe to anticipate that $H_{irr}(0)<10$ T, being much smaller than $H_{c2}(0)\approx 15$ T.

$$E=\frac{R}{2}\frac{d(\mu_0 H)}{dt} \quad (4)$$

and j_c can be determined from the width of the microhysteresis loops around a certain field via $j_c=20\Delta M/Va(1-a/3b)$ based on the assumption that the current density j_c is uniform throughout the cross section of the ring.[21] Since the vortices are forced to move by the external field sweeping, this process is thus called dynamical relaxation.[19,20] For a superconducting disk the magnetic moment is contributed mainly from the current circulating near the perimeter of the sample. For example, 7/8 of the total magnetic moment is contributed by the current flowing from $1/2R$ to R of a superconducting disk, where R is the radius of the disk. Therefore, from a rough estimation, it is safe to derive the $E(j)$ or $V(I)$ relation for a disk sample by using Eq. (4). As indicated in Ref. 20 and 19, the normalized relaxation rate can be determined via $Q=d\ln j_s/d\ln E$ in the dynamical relaxation process, and it should be identical to $S=-d\ln M/d\ln t$ determined in the conventional relaxation process. Later on Wen *et al.*,[22] Perkins and Caplin,[23] and Ji *et al.*[24] have shown that the conventional relaxation, dynamical relaxation, and the dc transport method should give the same information of flux motion although the voltage range is different (the voltage in the dynamical relaxation is much higher than that in the conventional relaxation).

The raw data with two different field sweeping rates (0.02 and 0.01 T/s) are shown in Fig. 2. The Q values vs field for different temperatures are determined and shown in Fig. 4. It is clear that the relaxation rate increases monotonically with external magnetic field and extrapolates to 100% at about the melting point H_{irr}. At 2 K it is found from the $Q(H)$ data that the melting field (where $Q=1$) is about 8.7 ± 0.5 T, being close to $H_{irr}(T=2\ \mathrm{K})\approx 9.2T\pm 0.5$ determined from the $j_c(H)$ curve. It is known that the $H_{irr}(T)$ is rather stable in the low-temperature region; therefore, we can anticipate a rather low value of $H_{irr}(0)$ which is below 10 T, being much lower than $H_{c2}(0)$. As already pointed out in our earlier publication,[6] the large separation between $H_{irr}^{bulk}(0)$ and $H_{c2}(0)$ may manifest the existence of a quantum vortex liquid due to a strong quantum fluctuation of vortices in the pure system of MgB_2.

Theoretically, quantum melting of the vortex solid has been proposed by some authors[25–27] and preliminarily verified by experiments.[28,29] Solid evidence is, however, still lacking mainly because either the values of $H_{irr}(0)$ and $H_0(T)$ are too high to be accessible, such as in the classical Chevrel phase PbMoS system,[30] or the separation between them is too small,[28,29] leading to a difficulty in drawing any unambiguous conclusions. Here we try to have a rough consideration on the quantum melting field H_m proposed by Blatter *et al.*[25] for a two-dimensional (2D) system,

$$H_m(0)/H_{c2}(0)=1-1.2\exp(-\pi^3 C_L^2 R_Q/4R_{2D}), \qquad (5)$$

where C_L is the Lindermann number, $R_Q=\hbar/e^2\approx 4.1\ \mathrm{k}\Omega$, and R_{2D} is the sheet resistance. Since the new MgB_2 sample has a much higher charge density and thus a much lower sheet resistivity, according to the above relation, H_m should be more close to $H_{c2}(0)$ compared to high-T_c superconductors (HTS's). This is in contrast to experimental observations which may be explained as that the MgB_2 is not a quasi-2D system. Another approach was proposed by Rozhkov and Stroud,[31]

$$H_m(0)/H_{c2}(0)=B_0/[B_0+H_{c2}(0)], \qquad (6)$$

with $B_0=\beta m_p C^2 s\Phi_0/4\pi\lambda(0)^2 q^2$, where s is the spacing between layers, m_p is the pair mass, q the pair charge ($\approx 2e$), C the light velocity, $\lambda(0)$ the penetration depth at zero K, and $\beta\approx 0.1$. If comparing again the present superconductor MgB_2 with HTS's, $\lambda(0)$, q and β are more or less on the same scale; the difference comes from m_p and s. Therefore a preliminary conclusion would be that in MgB_2 either the pair mass m_p or the layer spacing s is much smaller than that of HTS's.

B. Residual relaxation rate at 0 K and weak temperature dependence of the relaxation rate

A strong quantum fluctuation normally favors a strong quantum tunneling creep. In order to see that, we plot in Fig. 5 the temperature dependence of the relaxation rate Q. It is clear that there is a clear residual relaxation rate for all fields and the relaxation rate in the wide temperature region stays rather stable against thermal activation and fluctuation. According to the thermally activated flux motion model,

$$E=v_0 B\exp\left(-\frac{U(j_c,T)}{k_B T}\right), \qquad (7)$$

where E is the electric field due to TAFM over the activation energy $U(j_c,T)$, v_0 is the average velocity of the flux motion, and B is the magnetic induction. The relaxation rate is

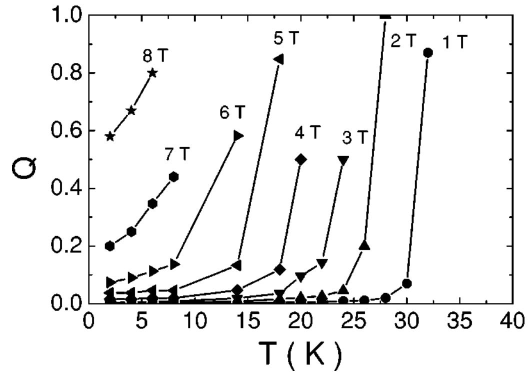

FIG. 5. Temperature dependence of the relaxation rate at fields of 1–8 T with increments of 1 T. A clear residual relaxation rate is observed at all magnetic fields. The relaxation is weakly dependent on temperature until the irreversibility temperature is reached. These are difficult to understand in the framework of thermal acitivation flux motion and thermal deepening.

$$Q=\frac{d\ln j_c}{d\ln E}=-k_B T\left(\frac{j_c dU(j_c,T,B)}{dj_c}\right). \qquad (8)$$

For any kind of $U(j_c)$ relation, a finite slope of dU/dj_c is expected. Therefore a much stronger temperature dependence of Q should be expected for thermally activated flux motion. This is in contrast to the experimental data. However, when the melting point $H_{irr}(T)$ is approached the relaxation rate will quickly jump to 100%. This may indicate that the thermal fluctuation is not the dominant process for the flux depinning in the superconductor MgB_2. It shows a high possibility for vortex quantum melting even at a finite temperature. Worthy to note is that the quantum tunneling rate is extremely low at an intermediate field, such as $Q=0.3\%$ at 2 K and 2 T, but is rather high at a high field, for example, $Q=20\%$ at 2 K and 7 T. This may imply that the field will greatly enhance the vortex quantum fluctuation and tunneling.

The small relaxation rate at a relatively low field has also been measured by Thompson *et al.*[5] who regarded it as a highly stable superconducting current density in MgB_2. Actually the relaxation rate can be rather high when the magnetic field is increased to a higher value. The extremely small relaxation rate and weak temperature dependence at a finite temperature at a low field is probably induced by a strong pinning barrier relative to the thermal energy, i.e., $k_B T \ll U_c$, where U_c is the intrinsic pinning energy. Recently it was concluded[32,33] that U_c is on the scale of 1000 K, being much higher than the thermal energy $k_B T$. Therefore for the superconductor MgB_2 the pinning well is too deep, leading to a trivial influence of the thermal activation and fluctuation. It thus naturally suggests that quantum fluctuation and tunneling play an more important role. Therefore, together with the fact discussed in last subsection, it is tempting to suggest that at a finite temperature the melting between a vortex solid and a liquid is due to quantum fluctuation instead of the thermal fluctuation.

V. CONCLUSION

In conclusion, in a thin film sample of MgB_2, the flux dynamics and the irreversibility field are investigated. Just like in bulk samples, it is found that the irreversibility field is rather low compared to the upper critical field in the low-temperature region, showing the possible existence of a quantum vortex liquid due to strong quantum fluctuation. The weak temperature dependence but strong field dependence of the relaxation rate may further suggest that the vortex melting at a finite temperature is also induced by the strong quantum fluctuation. The reason for such a strong quantum effect is still unknown, but it may be related to the superconducting mechanism of MgB_2, such as the relatively low upper critical field.

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