Flux Creep Relaxation in Single Crystal Hg-1201

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Abstract. Magnetization measurements on a single crystal of the high-Tc cuprate HgBa2CuO4 (Hg-1201) are performed for the two field orientations B||c and B||(a,b). Besides zero-field-cooling (zfc) – field-cooling (fc) and magnetization studies, the flux creep relaxation is determined. The zfc curves exhibit pronounced step-like anomalies in the mixed state above the lower critical field Bc1. They are interpreted as those points in the phase diagram where the penetrating flux fronts meet at the center of the sample. In order to obtain more information about the flux pinning mechanism flux creep relaxation experiments are performed at different temperatures in magnetic fields of 0.15 and 2 T. The resulting supercurrent dependence on the mean activation energy is analysed according to the collective pinning theory which predicts $U \propto \langle |j|^m \rangle - 1$. The $\mu$-values for B||c are 0.5 at higher temperatures and 2 at 5 K, and they vary between 1.1 and 1.5 for B||(a,b). The observed behaviour is quite different from that found for powder samples.

1. Introduction
The cuprate HgBa2CuO4 (Hg-1201) was first synthesized by Putilin et al. [1] and belongs to the family of mercury-based high-Tc superconductors with the record transition temperature of 135 K. Hg-1201 is the most simple representative of this class and in some sense it plays the role of a model system [2, 3]. It has tetragonal symmetry, with one single CuO-layer per unit cell, and becomes superconducting at about 96 K at optimal doping. Its relative simplicity and potential for applications make Hg-1201 a desirable system for the investigation of vortex dynamics and pinning properties.

However, only few investigations on single crystals of the Hg-based compounds exist up to now, because sizable crystals have not been available. Recently, we conducted a comparative study of the ac and dc magnetization of a Hg-1201 single crystal of about 100 mm³ volume [4]. We found that the second peak in the imaginary part of the ac susceptibility, characteristic of Hg-1201 powder samples, is shifted to higher temperatures. This indicates that the transition between different types of vortex pinning is shifted to higher magnetic fields and temperatures in single crystals.

Nevertheless, step-like anomalies in the temperature dependence of the zfc curves were observed. They occur at around 35 K in both powders [5] and single crystals [4]. In addition, for the orientation B||(a,b), a second step occurs at around 10 K, reflecting a clear orientation dependence of this anomaly. The origin of this anomaly is not
yet clear. Different models were considered, i.e., crossing with the lower critical field \[6\] or temperature dependent changes in vortex mobility \[5\].

In order to obtain information about possible pinning mechanisms, we perform a detailed study of a Hg-1201 single crystal for two magnetic field orientations, parallel to the \(c\)-axis and parallel to the \(ab\)-planes. Experimental data for the zfc-fc behaviour, and especially flux creep relaxation measurements of the magnetic moment, already applied to powder samples \[5\], are evaluated. The analysis results in a clear geometry dependence which determines the flux distribution inside the sample governed by the critical current density.

2. Experimental

Single crystals of HgBa\(_2\)CuO\(_{4.4}\) were grown following the procedure described in Ref. \[2\]. For the present study, we used a small single crystal with a mass of 0.69 mg and dimensions of about 0.19 mm along the \(c\)-direction and 1.1 x 0.48 mm\(^2\) in the \(ab\)-plane. Magnetization and flux creep relaxation measurements are performed by means of a commercially available magnetometer (MPMS, Quantum Design Inc.).

3. Results and discussion

Figures 1 and 2 show the temperature dependence of the magnetic moment after zero field cooling for fields parallel to the \(c\)-axis and the \(ab\)-plane, respectively. Measurements are performed in different fields, as indicated. The zfc curves allow an estimation of the lower critical field \(B_{c1}(T)\): below \(B_{c1}\) the temperature dependence of the magnetic moment should be rather weak and only depend on the geometry which brings the sample into the intermediate state. However, when crossing \(B_{c1}\), flux penetrates into the sample and the magnetic moment starts to approach zero. The resulting values are plotted in Fig. 3, which shows the phase diagram. The phase boundaries agree with those determined from the first deviation from linearity in the magnetization curves. All magnetization curves for \(B || c\) exhibit a pronounced peak (“fishtail effect”) as also observed for single crystals of other high-\(T_c\) cuprates \[7\]. Due to limited space, this topic is not discussed here.

As can be seen from Fig. 3, \(B_{c1}\) is larger by a factor of about 3 for \(B || c\) compared to \(B || (a,b)\), a value also found earlier for Y-Ba-Cu-O crystals \[8\]. For both orientations the temperature dependence is linear, as expected from a Ginzburg-Landau phase transition, below 0.5 \(T_c\). Following the lines in Fig. 3 indicating the zfc measurements suggests an interpretation based on the Bean-critical-state model, which was already proposed for Y-Ba-Cu-O crystals \[8\]. With increasing temperature flux starts to penetrate the sample after passing the lower critical field \(B_{c1}\), causing the steep reduction of the magnetic moment \(M\). Once the flux fronts meet at the center of the sample, further reduction of \(M\) slows down, which generates the step-like anomalies in the zfc curves. For \(B || c\), this kink occurs at 23 K in a field of 100 mT (above \(B_{c1}\) at \(T = 0\)), but is shifted to 52 K in a field of 10 mT, because flux penetration only starts after crossing the \(B_{c1}\)-curve at 35 K. For the other orientation with \(B || (a,b)\), the flux fronts meet at lower temperatures due to the smaller sample size in \(c\)-direction. Flux penetration starts immediately when warming up the sample in fields of 10 and 100 mT, but is shifted to 61 K for warming up at 3 mT. Such an interpretation is also supported by results of the relaxation measurements.

![Figure 1](image1.png) **Figure 1.** Temperature dependence of the magnetic moment (normalized to the value at \(T = 4\) K) after zero field cooling (zfc) for several magnetic fields parallel to the \(c\)-axis (\(B || c\)).

![Figure 2](image2.png) **Figure 2.** Temperature dependence of the magnetic moment (normalized to the value at \(T = 4\) K) after zero field cooling (zfc) for several magnetic fields parallel to the \(ab\)-plane (\(B || (a,b)\)).
Flux creep relaxation measurements are performed for $B \parallel c$ and $B \parallel (a,b)$ at five temperatures ($T = 5, 10, 15, 20,$ and $25 \text{ K}$) after applying a magnetic field of 150 mT. Examples are shown in Fig. 4. Additionally for the orientation $B \parallel c$, relaxation measurements are performed after applying a much higher field of 2 T. The corresponding field and temperature values are marked in the phase diagram in Fig. 3. From this, it is clear that all relaxation processes take place in the mixed state, well above $B_{c1}$, but also well below the irreversibility field (e.g., 1 T at 80 K for $B \parallel c$ [4]). For both orientations the relaxation shows the logarithmic time dependence predicted by the Kim-Anderson model. The time dependence is described by $M(t) = M_0 - M_1 \ln(t/t_0)$, with $t_0 = 10^{-13}$ s for $B \parallel c$ and 15 s for $B \parallel (a,b)$ in our case. It should be mentioned that for much longer measurement periods deviations from logarithmic time behavior occur as already reported for YBa$_2$Cu$_3$O$_7$ single crystals [9]. For the following first analysis, however, this is not yet evaluated and taken into account.

The resulting flux creep rates $S = \frac{dM}{dt}/\ln(t)$ are plotted in Fig. 5. A considerable difference exists between the two orientations. For the orientation $B \parallel c$ the rate values are larger by more of a factor 100 compared to those for $B \parallel (a,b)$.

Figure 3. Phase diagram $B(T)$. Solid curves: lower critical field $B_{c1}(T)$. Dashed lines: step-like anomalies in the zfc-curves (square dots: $B \parallel (a,b)$, circular dots: $B \parallel c$). Horizontal lines: fields for zfc measurements. Triangles: fields and temperatures of relaxation measurements.

Figure 4. Some relaxation curves of the magnetic moment recorded after application of a magnetic field of 150 mT. The curves are normalized at the time 100 s. They follow a logarithmic time dependence (solid lines).

Figure 5. Temperature dependence of flux creep rates determined from relaxation measurements. The values for the orientation $B \parallel (a,b)$ are enlarged by a factor of 100 (solid lines are guides for the eyes).

Figure 6. Mean activation energy as a function of supercurrent density normalized to the critical current density for a magnetic field of 150 mT parallel the c-axis applied after zero field cooling.
the orientation $B \parallel (a,b)$. This corresponds roughly with the absolute values of the magnetization curves. For the orientation $B \parallel c$, however, a clear temperature dependence is observed. Both applied field values show an increase with decreasing temperature. The only exception occurs below 10 K for the field 150 mT, where the creep rate decreases considerably. This behavior might reflect the different flux penetration after the flux front meet in the center inside the sample that is also responsible for the step-like anomalies in the zfc curves. A corresponding change is observed in the current density dependence of the mean activation energy $U(j)$ (Fig. 6), where the behavior at 5 K also differs from that at higher temperatures.

All results of the relaxation measurements are summarized in Figs. 6 – 8. Following a method proposed by Maley et al. [10], the relaxation at different temperatures leads to a consistent plot of the activation energy $U$. It is based on the rate equation $dM/dt \propto \exp\{-U/kT\}$, leading to $U = kT(c - \ln(dM/dt))$. Here, $c$ is assumed to be constant and used as fitting parameter, yielding a smooth dependence of $U$ vs $M$. Since $M \propto j$, at least the functional form of $U(j)$ can be determined. In our case, this leads to a power law according to the collective pinning theory [11], which generally predicts $U \propto (j/j_c)^\mu - 1$ (solid lines in Figs. 6 – 8); $j_c$ is the critical current density and $\mu$ is a critical exponent that depends on dimensionality and effective bundle size of the vortex assembly.

For $B \parallel c$ and 150 mT we found $\mu = 0.5$ for higher temperatures and $\mu = 2$ for 5 K. $\mu$-values of this order were also reported for YBa$_2$Cu$_3$O$_7$ single crystals [9]. The value 2 is rather large and might indicate that the experimental conditions do not satisfy the theoretical requirements in this low-temperature region. In any case, however, there is no hint for single vortex creep, which requires a $\mu$-value as low as 1/7 = 0.14. The relaxation after applying the higher field of 2 T is similar (Fig. 7), the $\mu$-value for lower temperatures (5 and 10 K; $\mu = 1.3$) is larger than that for higher temperatures, $\mu = 0.17$. Both, however, are clearly reduced, the low-temperature value now close to the one for collective creep of small bundles, $\mu = 1.5$ [12]. For higher temperatures the $\mu$-values decrease, qualitatively similar to the behavior already reported for YBCO [9] and also predicted theoretically. For $B \parallel (a,b)$, there is less of a difference: $\mu = 1.5$ for higher temperatures and $\mu = 1.1$ for T = 5 K. The corresponding supercurrents, however, are smaller by a factor of more than 100.

Concluding these relaxation results, it seems that vortex bundle pinning is dominant within the whole region of the mixed state, with no indication of a transition between regimes of different type of vortex pinning as observed earlier in powder samples [13] where it might be due to surface effects. There is no evidence for single vortex pinning at low temperatures. The $\mu$-values at 5 K are rather large compared to those found for powder samples. The reason for this strong enhancement is not yet clear and might be connected with the special shape of the magnetization curves (fishtail effect). A further detailed relaxation investigation is necessary.

4. Conclusions
In contrast to earlier measurements on Hg-1201 powder samples no indication of a transition between different types of vortex pinning is found in single crystals of this superconducting cuprate system. This can be concluded
both from zfc-fc measurements and from flux creep relaxation experiments. The interpretation of the range of μ-values is not quite clear as yet, but the results indicate a more-or-less homogeneous pinning behavior in the mixed state. The flux distribution inside the sample seems to follow the Bean-critical-state model which allows an interpretation of the step-like anomalies of the zfc curves. This observation should be helpful for further studies concerning the connection of relaxation behavior and special characteristics of magnetization curves.

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References
[1] Putilin S N, Antipov E V, Chmaissem O and Marezio M 1993 Nature 362 226
[2] Zhao X, Yu G, Cho Y-C, Chabot-Couture G, Barisic N, Bourges, P, Kaneko N, Li Y, Lu L, Motoyama E M, Vajk O P and Greven N 2006 Adv. Mater. 18 3243
[3] Barisic N, Li Y, Zhao X, Cho Y-C, Chabot-Couture G, Yu G and Greven M 2008 Phys. Rev. B 78 054518
[4] Baenitz M, Lueders K, Maurer D, Barisic N, Cho Y, Li Y, Yu G, Zhao, X and Greven M 2009 doi:10.1016/j.physc.2009.05.203
[5] Maurer D, Breitzeke H, Lueders K, Baenitz M, Pavlov D A and Antipov E V 2005 Physica C 432 250
[6] Pissas M, Moraitakis E, Kallias G, Terzis A, Niarchos D and Charalambous M 1998 Phys. Rev. B 58 9536
[7] Werner M, Sauerzopf F M, Weber H W and Wisniewski A 2000 Phys. Rev. B 61 14795
[8] Kruzin-Elbaum L, Malozemoff A P, Yeshurun Y, Cronemeyer D C and Holtzberg F 1989 Phys. Rev. B 39 2936
[9] Thompson J R, Yang Ren Sun and Holtzberg F 1991 Phys. Rev. B 44 458
[10] Maley M P, Willis J O, Lessure H and McHenry M E 1990 Phys. Rev. B 42 2639
[11] Blatter G, Feigel’man M V, Geshkenbein V B, Larkin A I and Vinokur V M 1994 Rev. Mod. Phys. 66 1125
[12] Yeshurun Y, Malozemoff A P and Shaulov A 1996 Rev. Mod. Phys. 68 911
[13] Maurer D, Lueders K, Baenitz M, Pavlov D A and Antipov E V 2008 Physica C 468 1305