Thermally activated flux creep in K_3C_{60} single crystals

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Long-term magnetic relaxation in the fullerene superconductor K_3C_{60} was measured on crystalline samples in a wide range of temperatures and magnetic fields. A logarithmic $M(t)$ dependence is observed on single crystals with 100% shielding fraction. The relaxation rate at different magnetic fields increases progressively with temperature. The flux-creep activation energy is found to be in the range from 10 to 80 meV with a peak in its temperature dependence. From measurements on samples with nonperfect stoichiometry we show that inhomogeneities strongly affect the relaxation process and may mask a logarithmic behavior. [S0163-1829(96)07045-2]

One of the most remarkable features of type-II superconductors regarding the dynamics of flux motion is the relaxation of the magnetization $M(t)$ at fixed temperature T and magnetic field B. The relaxation process has been the subject of intensive studies because it heavily affects the current-carrying capability of high temperature superconductors. Commonly, the relaxation is described by the flux creep activation energy U_0. The Anderson-Kim model[1] assumes a uniform barrier U_0 for the depinning of vortex bundles, leading to a logarithmic time dependence of the magnetization:

$$M(t)=M_0\left[1-\frac{kT}{U_0}\ln\left(\frac{t}{t_0}\right)\right], \tag{1}$$

where M_0 is the unrelaxed value of M and t_0 is a time constant. Investigations of the relaxation in high-T_c superconductors performed by magnetic measurements demonstrated that this logarithmic dependence was observed in most cases[2–9] and resulted in an activation energy U_0, which initially increases with increasing temperature and then decreases after having reached a maximum at some temperature T_m.[3,4,6–8,10] A number of theoretical explanations for this phenomenon were proposed.[4–6,11–13]

Fullerene superconductors show similar properties as the cuprates, e.g., relatively high values of the critical temperature and of the Ginzburg-Landau parameter (for reviews see Refs. 14–16). Also, significant relaxation can be found in fullerene superconductors. It is interesting to measure the time dependence of this process and to compare it to the high T_c's. However, although several years have passed since the discovery of superconductivity in fullerenes,[17] only a few results on flux pinning[18,19] and magnetic relaxation[20–23] were published. In Ref. 20, the flux creep activation energy was estimated to be of the order of 10^{-2} eV. However, it should be pointed out that all of the previous measurements had to be performed on powder samples, where the magnetic relaxation usually did not show a logarithmic time dependence.[21] Even some peaks were observed in $M(t)$ curves during short-term relaxation.[23] This behavior could be connected to an intergranular interaction between grains in powder samples as well as to weak links, which may exist in samples of a poor quality. These factors may strongly affect the relaxation process.

In this work we present results on magnetic relaxation obtained in crystalline fullerene superconductors. The measurements were done on samples of different quality. Our experiments show that instabilities of the vortex system completely mask the logarithmic process in the presence of impurities and weak links in the sample. Magnetic relaxation in a crystalline sample of good quality shows clearly a logarithmic behavior of the relaxation and allows us to calculate the relaxation rate S and the flux creep activation energy U_0 in a wide range of temperatures and fields. We obtain U_0 values between 10 and 80 meV. The temperature dependence of the activation energy shows a smooth peak.

The main task of this work was to investigate the long-term magnetic relaxation in a sample of a good quality, in order to avoid influences of intergranular currents. For this purpose our measurements were performed on crystalline K_3C_{60} samples with 100% shielding fraction. Additionally we measured crystalline specimens with different values of the superconducting fraction (from 25% to 100%) in order to investigate the influence of nonsuperconducting inhomogeneities.

Three stoichiometric reactions of K_3C_{60} were performed in evacuated pyrex tubes, using potassium azide KN_3 as a

convenient source of potassium, which can be accurately weighed in air in small quantities. The parameters of the reactions were the following.

(1) A set of four C_{60} single crystals with a total mass of 11.12 mg was reacted with 3.85 mg KN_3 at 500 °C for 44 h. The final total mass after the reaction was 13.2 mg. dc superconducting quantum interference device (SQUID) measurements on four crystals (sample S1) gave $T_c=19.4$ K with 100% shielding and 9% Meissner fraction. Two of these crystals were then sealed separately in quartz tubes. One was analyzed by x-ray diffraction and exhibited the appropriate relative intensities of [111], [222], and [333] reflections for single phase K_3C_{60} and no evidence for other phases such as KC_{60} or C_{60}. The mosaic spread was 3° full width at half maximum (FWHM), much worse than that of a parent crystal from the same batch. The second of these crystals (S2, 2.1 mg) was remeasured with a dc SQUID and showed $T_c=19.2$ K with 100% shielding and 10% Meissner fraction. Preliminary ac measurements on sample S2 did not show any sign of granularity for superconducting current flow.

(2) Two C_{60} crystals with a total mass of 1.8 mg were reacted with 0.7 mg KN_3 and heated to 500 °C for 46 h. The final total mass after the reaction was 1.8 mg (we presume that some amount of C_{60} sublimated during reaction). Preliminary dc SQUID characterization of both crystals together gave $T_c=19.2$ K with 18% shielding and 6% Meissner fraction. One of these crystals was then sealed in quartz for Raman measurement (sample S3) which showed a spectrum characteristic of amorphous carbon and a very weak Ag(2) mode at the undoped C_{60} position. Neutron diffraction on the same crystal revealed a poor mosaic structure (5° FWHM) and an intensity ratio 220/311$\sim$1.3, intermediate between the expected 1.8 and 0.64 for C_{60} and K_3C_{60}, respectively. There is no doubt that this crystal contained some K_3C_{60} and a potassium-deficient compound (pure C_{60} or K_1C_{60}), dc SQUID measurement on this sample showed 25% shielding fraction.

(3) One large C_{60} crystal (11.73 mg) was reacted with 4.12 mg KN_3. The final total mass after the reaction was 13.5 mg. The crystal was then broken into six pieces and annealed at 360 °C for 44 h to improve the homogeneity. An x-ray contour plot of one crystal in the region of the (111) reflection showed a large C_{60} peak with a narrow mosaic and weaker K_3C_{60} peaks with broad mosaic spread. One piece (4 mg) was then sealed separately (sample S4) and dc SQUID measurements showed $T_c=19.5$ K with 65% shielding and 7% Meissner fraction.

Magnetic dc measurements on S1, S2, S3, and S4 were performed in a commercial SQUID magnetometer. The temperature range of these measurements extended from 5 K to T_c, the range of magnetic fields was $\mu_0 H_{c1} \ll 0.1\ \mathrm{T} \leqslant \mu_0 H \leqslant 1\ \mathrm{T} \ll \mu_0 H_{\mathrm{irr}}$. This range of external fields ensured complete flux penetration into the samples. The ramp rate of the magnetic field in our experiments was of the order of 28 mT/s and the same for all applied fields in our experimental window. The magnetic relaxation was recorded up to 5×10^4 s. The measurements were performed as follows. The specimen was cooled from $T=30\ \mathrm{K} > T_c$ down to $T<T_c$ in zero external field. After temperature stabilization, an external field (H_{ext}) was applied. The magnetization was monitored at fixed T and H_{ext}. The first measurements were performed at $t_0\sim80$ s after field stabilization. Consecutive measurements were carried out every 60–63 s. The data obtained on S1 and S2 are very similar. In this paper mainly results on sample S1 are shown.

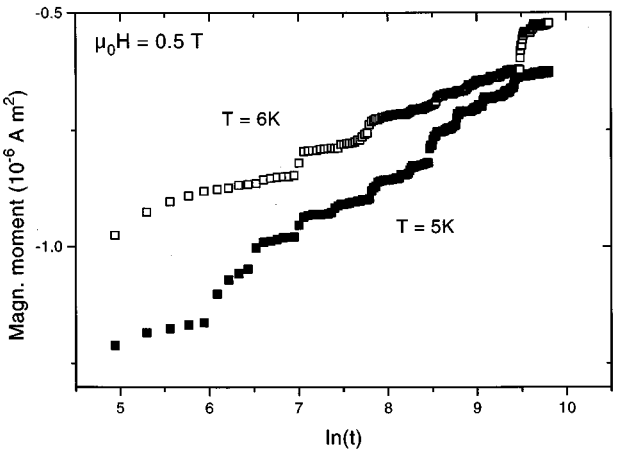

FIG. 1. Time dependent magnetic moment of sample S3 in an external magnetic field $\mu_0 H_{\mathrm{ext}}=0.5$ T at 5 K (solid symbols) and at 6 K (open symbols).

The magnetic relaxation measured on samples S3 and S4 with nonperfect shielding is presented in Fig. 1. The relaxation did not show a logarithmic behavior and jumps of the magnetization appeared. In order to check for possible instabilities of our experimental system, the same measurements were performed in another noncommercial SQUID magnetometer. While most of the small jumps in $M(t)$ could not be observed in this magnetometer because of its worse sensitivity, the big ones were clearly seen. Additionally, a single crystal of Bi-2212 was measured in the commercial magnetometer, which showed a very smooth logarithmic relaxation. Therefore, we conclude that the unusual magnetic relaxation shown in Fig. 1 is an intrinsic property of samples with nonideal shielding fraction. We believe that the presence of such imperfections in powdered samples has also led to the nonlogarithmic relaxation reported in Refs. 21–23. The jumps can be explained if we assume that these samples have a nonideal stoichiometry and that superconducting regions are surrounded or separated by nonsuperconducting ones in the specimens. In this case, the diffusion of magnetic flux may be more difficult because of a mismatch between neighboring regions or a disturbance of the structure at the surface of grains or simply by impurity phases between the grain boundaries. All of these imperfections can strongly affect the relaxation process and the expected logarithmic behavior will be partly or completely masked.

The magnetic relaxation obtained on samples S1 and S2 with 100% shielding fraction is completely different in character. The time-dependent magnetization of sample S1 at an external magnetic field, $\mu_0 H_{\mathrm{ext}}=0.1$ T and for temperatures from 5 to 17 K is presented in Fig. 2. All the results obtained on sample S2 are in a very good agreement with those shown for S1. The results follow the logarithmic expression (1). The temperature dependence of the creep rate $\delta M(t)/\delta \ln t$ at different values of the magnetic field is shown in Fig. 3. The creep rate decreases linearly with increasing temperature and

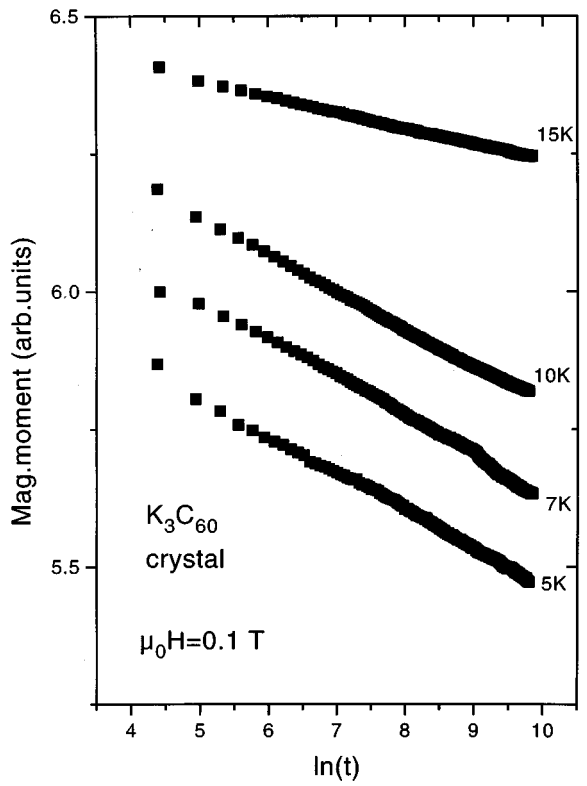

FIG. 2. Time dependent magnetic moment of sample S1 in an external magnetic field $\mu_0 H_{\text{ext}}$=0.1 T at 5, 7, 10, and 15 K.

extrapolates to zero at some temperature $T_0 \sim 18.1$ K$<T_c$, which is the same for all external fields within our experimental window.

The temperature-dependent activation energies are obtained from the decay of the magnetization with the relations[24]

$$U_0 = \frac{k_b T}{S}; \quad S = \frac{1}{M_0}\frac{\delta M}{\delta \ln t}, \tag{2}$$

where S is the normalized relaxation rate and M_0 is the initial value of the magnetization after the external field was applied. For estimating the relaxation rate S and the flux creep activation energy U_0, we use the value of the first recorded magnetization $M(t_0)$ as M_0 in Eq. (2). The temperature dependences of S and U_0 at different applied fields are shown in Figs. 4 and 5, respectively. The relaxation rate increases smoothly with increasing temperature at all fields and becomes larger with increasing external field. Thus our results contradict an earlier report,[22] according to which the relaxation rate in K_3C_{60} displays either a broad maximum or a plateau. This could be due to the fact that a powdered sample was measured in Ref. 22.

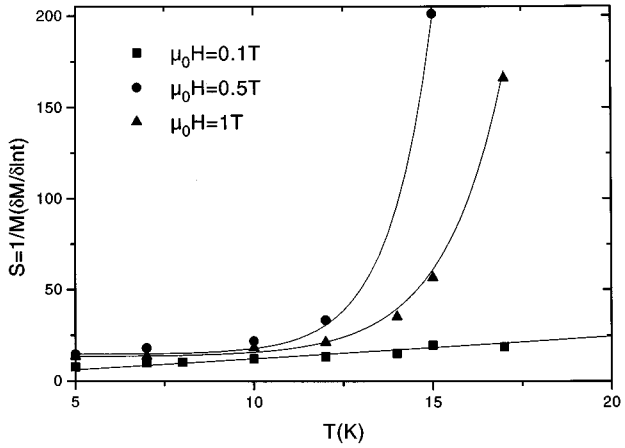

FIG. 4. Temperature dependence of the normalized relaxation rate at 0.1, 0.5, and 1.0 T.

The flux creep activation energy, shown in Fig. 5, first increases with temperature and then reaches a peak at some temperature T_m. This temperature T_m decreases almost linearly with increasing external field. Thus the temperature dependence of U_0 is similar to that observed in high-T_c superconductors.

In conclusion, the relaxation rate and the activation energy U_0 for flux creep in K_3C_{60} crystals were studied experimentally. It was shown that inhomogeneities in a superconductor strongly affect the relaxation process and may mask a logarithmic dependence, while for good quality samples the magnetic relaxation follows the $M \sim \ln t$ behavior. We observe that the flux-creep rate increases progressively with temperature, at least up to $T=17$ K$\simeq 0.88T_c$. The temperature dependence of the flux-creep activation energy exhibits

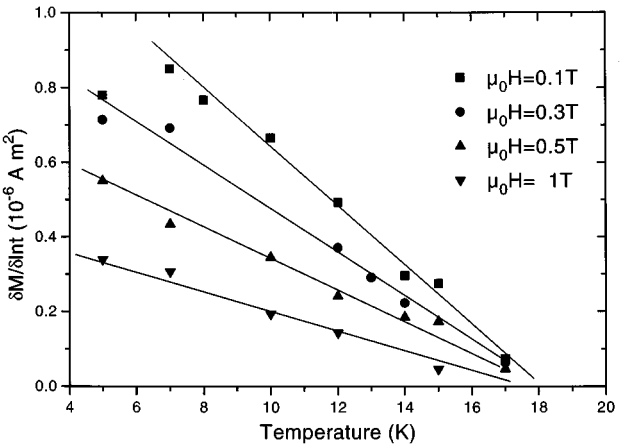

FIG. 3. Temperature dependence of the creep rate, $\delta M(t)/\delta \ln t$, at different magnetic fields.

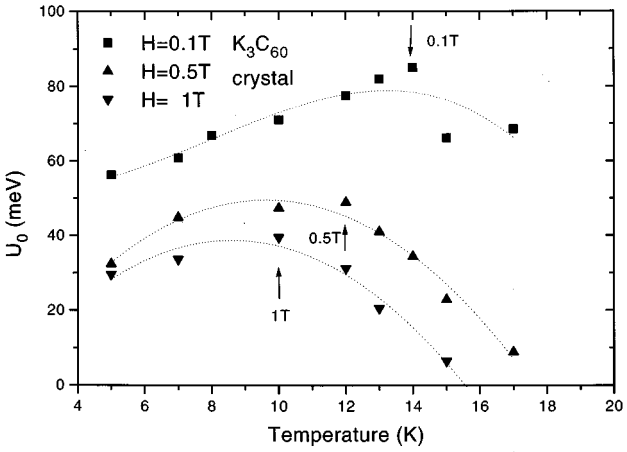

FIG. 5. Temperature dependence of the flux creep activation energy calculated from Eq. (2).

a peak at some characteristic temperature T_m, which depends roughly linearly on the magnetic field.

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