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Low-field magnetic-relaxation effects in $CeCu_2Si_2$

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We have found time effects in the magnetization M of the heavy-fermion $CeCu_2Si_2$ at $H<400$ Oe similar to the effects we recently reported for the high-T_c superconductors Sr-La-Cu-O and Ba-La-Cu-O. If a small field H_i is applied to a virgin *bulk* polycrystalline specimen, the magnetization M relaxes with time accurately following the law $M(t)-M(t_0)\propto\ln(t/t_0)$ over observation times of 10^5 sec. The same law is followed at $H=0$ when the field H_i is removed. For $H<H_{c1}$ the logarithmic rate $\partial M/\partial\ln t$ is directly proportional to H_i^4 and becomes almost independent of H_i when flux penetrates into the sample.

In a recent paper[1] we presented a set of new relaxation effects observed in the high-T_c superconductors Ba-La-Cu-O and Sr-La-Cu-O at $T\ll T_c$. These showed that the magnetization was unstable at $H<H_{c1}$ and decayed in time following a logarithmic law. We recently looked for such an effect in the classical superconductors Nb, Ta, Nb-Ti, Pb-In without finding any relaxation with a logarithmic time dependence for $H<H_{c1}$. We have, however, discovered relaxation effects with logarithmic time dependences in a different superconducting material not belonging to the group of new high-T_c superconducting oxides. This material is the heavy-fermion superconductor $CeCu_2Si_2$ with a transition temperature $T_c=0.6$ K. In this case the effects were observed in *bulk* polycrystalline specimens as well as in powdered specimens.

The first observation of metastable states in the magnetization of Ba-La-Cu-O had been reported by Müller, Takashige, and Bednorz.[2] Switching off the field after cooling in a field or switching on a field after zero-field cooling created metastable states which decayed exponentially for short times and later on at a considerably slower rate. More recently, Giovannella, Collin, Rouault, and Campbell[3] have studied vortex creep in La-Sr-Cu-O using torque measurements. For moderate applied fields they obtained decays which were nonexponential and tended toward a power-law form.

In our study of Sr-La-Cu-O and Ba-La-Cu-O at low fields ($0<H<385$ Oe) we found that, following a small increase or decrease of the magnetic field, the isothermal dc magnetization M decayed in time as $M\propto\ln(t/t_0)$ with great accuracy over observation time as long as 10^5 sec. Furthermore, the logarithmic decay rate at $H=0$ could be expressed as

$$\frac{\partial M}{\partial\ln t}\propto TH_i^3\,, \qquad (1)$$

for $1<T<9$ K and $20<H_i<385$ Oe. Here H_i is the external field which is applied and reduced to zero before the decay of the magnetization is measured.

One problem in interpreting the results described by expression (1) was caused by the granular nature of our oxide specimens. Both sintered and powdered specimens showed irreversible increases in the isothermal dc susceptibility at very low fields,[4] indicating a continuous distribution of shielding currents across *different* grains joined by weak Josephson junctions. The resulting *intergrain* superconducting glass behavior could be reduced considerably after powdering the specimen but it was still noticeable in the dc susceptibility.

Our experimental arrangement for measuring isothermal dc magnetization using noncommercial superconducting quantum interference device magnetometers has already been described in Ref. 1. Figure 1 shows the decay of the dc magnetization in arbitrary units as a function of the logarithm of the time. The specimen is bulk polycrystalline $CeCu_2Si_2$ in the form of a parallelepiped ($1.1\times2.1\times6.5$ mm^3). In this measurement, after zero-field-cooling the sample, an external field is applied and immediately again reduced to $H=0$ at $t=t_0$. We notice that M very accurately follows the law

$$M(t)-M(t_0)\propto\ln(t/t_0)\,, \qquad (2)$$

over an observation time of four decades. This law and the slope of M vs $\ln t$ do not depend on waiting times at $H=H_i$. A similar $\ln t$ law is obtained if the field is raised from $H=0$ to H_i and the decay measured at $H=H_i$. In this case M decays in the opposite direction, that is, the absolute value of M decreases as a function of time or the specimen becomes less diamagnetic.

It is well known that in type-II superconductors at the critical state, vortices move according to the Anderson-Kim thermally activated flux-creep process.[5,6] The total flux ϕ in a cylindrical sample of radius R in a longitudinal field, at the time t given by[7]

$$\phi(t)=\phi(t_0)\pm\frac{\pi}{3}k_BTR^3\left(\frac{\partial U_{\rm eff}}{\partial|\nabla B|}\right)^{-1}\ln(t/t_0)(1+\delta)\,, \qquad (3)$$

where $(\partial U_{\rm eff}/\partial|\nabla B|)$ is the change in the activation energy $U_{\rm eff}$ with the flux density gradient. δ is a term which is small compared to unity.

Expression (3) was confirmed by Beasley, Labusch, and Webb[7] in experiments with Pb-Tl alloys for $H>H_{c1}$. It is important to note that, in those experiments done in

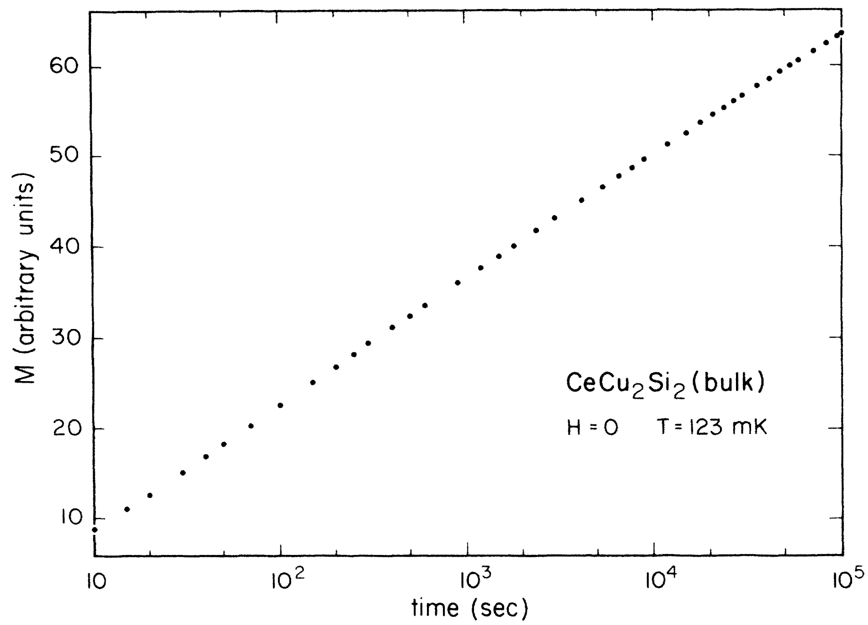

FIG. 1. Decay of the magnetization M at $H=0$ as a function of the logarithm of time for bulk polycrystalline $CeCu_2Si_2$ at $T=123$ mK after a field $H_i=200$ Oe has been turned off.

magnetic fields $H<H_{c1}$, *no flux creep* was observed. Above H_{c1} the logarithmic creep rate showed a modest value almost independent of the applied field H. As H_{c2} was approached the flux creep rate showed a considerable increase.

Flux creep following a rather strong logarithmic time dependence ($\simeq$9% decade) has recently been observed at $H=1$ T in a single crystal of $Y_1Ba_2Cu_3O_{7-x}$ by Worthington, Gallagher, Dinger, and Sandstrom.[8]

Contrary to previously known cases of flux creep in type-II superconductors, the logarithmic time rates observed by us at $H<H_{c1}$ do not seem to depend directly on flux density gradients. This is shown in Fig. 2. Here we have plotted the absolute value of $\partial M/\partial \ln t$ at $T=25$ mK as a function of the field H_i for the powdered $CeCu_2Si_2$ specimen. Each point in this curve is derived from a set of data of the kind shown in Fig. 1. The closed circles correspond to measurements at $H=H_i$ after the field had been raised from $H=0$ in the virgin specimen and the open circles correspond to measurements at $H=0$ after the field had been reduced from $H=H_i$. The H_i^4 line is drawn as a guide to the eye. We notice that both sets of data follow the same behavior as a function of H_i with the field-on data (solid circles) about a factor of 3 larger than the $H=0$ data. Depending on the value of H_i, this specimen showed only some few percent of nonreversible behavior of the magnetization after field cycling to H_i. In consequence the flux density gradients left at $H=0$ from the irreversible behavior were much too small in comparison to the flux density gradients created with the field on to account for the factor of 3 between the relaxation rates observed.

In Fig. 2 we see that for fields $10<H_i<50$ Oe the logarithmic rate approximately follows

$$\partial M/\partial \ln t \propto H_i^4 . \quad (4)$$

This dependence is stronger than that found for Sr-La-Cu-O [expression (1)]. The leveling off of $\partial M/\partial \ln t$ at higher fields follows the behavior of the dc magnetization of $CeCu_2Si_2$ as given by Rauchschwalbe *et al.*[9]

In Fig. 3 we compare the logarithmic rate at $H=0$ and $T=123$ mK in two different specimens. The filled trian-

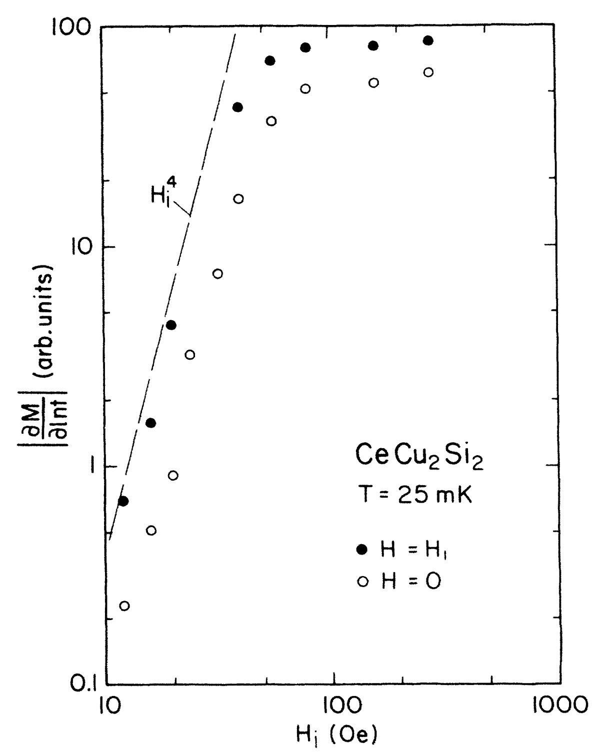

FIG. 2. Absolute value of the logarithmic decay rate at $T=25$ mK as function of the field H_i.

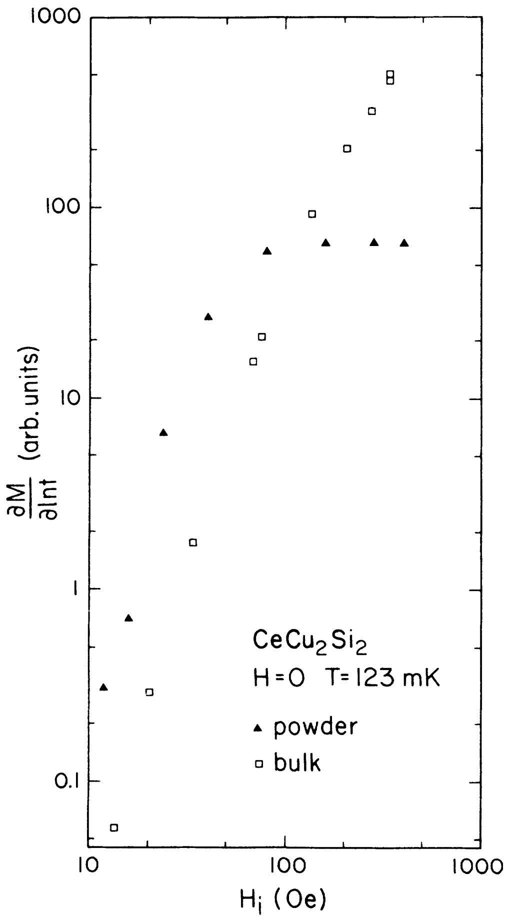

FIG. 3. Logarithmic decay rate at $H=0$ as a function of the field H_i. ▲ correspond to powdered $CeCu_2Si_2$, and □ correspond to bulk $CeCu_2Si_2$.

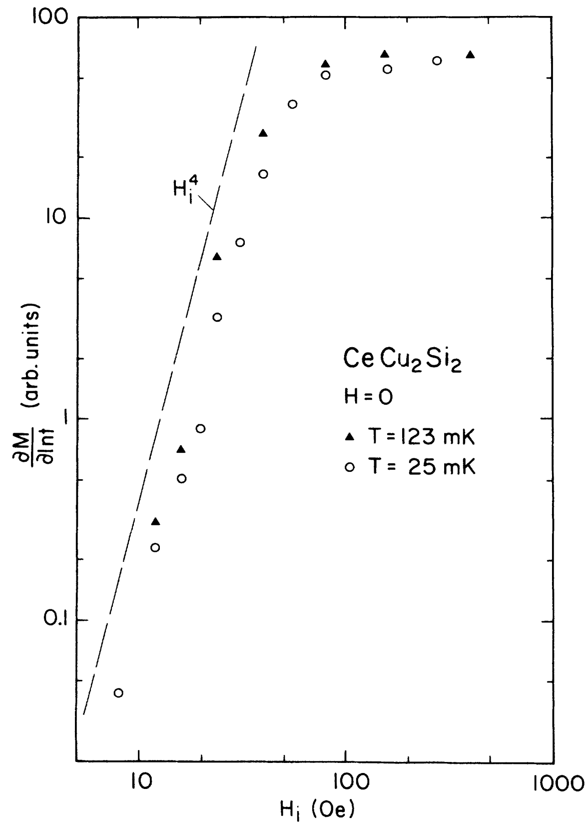

FIG. 4. Logarithmic decay rate at $H=0$ for powdered $CeCu_2Si_2$ at two different temperatures.

gles correspond to powdered $CeCu_2Si_2$ and the open squares to the bulk polycrystalline specimen. At the lowest fields we find $\partial M/\partial \ln t \propto H_i^4$ for both specimens with a coefficient about 10 times larger for the powdered specimen than for the bulk material. Furthermore, the leveling off of $\partial M/\partial \ln t$ from the H_i^4 law found in the powdered samples is not seen in the bulk material and must occur at higher fields. This is to be expected in view of the different demagnetization factors in the two specimens.

The temperature dependence of the logarithmic relaxation rate was also investigated. In our previous work on Sr-La-Cu-O we found that between $T=1$ and 9 K the rate $\partial M/\partial \ln t$ increased by about a factor of 10, indicating that the relaxation rate was thermally activated. Below 1 K, for Sr-La-Cu-O, the relaxation rate entered into a *temperature-independent* regime indicating that at low temperatures a different mechanism drives the relaxation of the magnetization M. In the present measurements we also observed relaxation rates which are almost independent of temperature as shown in Fig. 4 for powdered $CeCu_2Si_2$ ($T_c=0.6$ K). Here the open circles correspond to data at $T=25$ mK and the closed triangles to $T=123$ mK. We see that a change in temperature by a factor of 5 produces only a change in $\partial M/\partial \ln t$ by a factor of 1.5.

The relaxation rate for $CeCu_2Si_2$ was found to be $\Delta M/M \simeq 0.4\%$ per decade of time in seconds at $T/T_c=0.21$ and $H=40$ Oe. This value is of the same order of magnitude as the rate found for Sr-La-Cu-O: $\Delta M/M=0.1\%$ per second decade at $T/T_c=0.26$ and $H=40$ Oe.[10] In both expressions M is the magnetization of the corresponding material at $H=40$ Oe. The results seem to indicate that problems associated with sample preparation, which are known to affect the superconducting properties of $CeCu_2Si_2$ (Ref. 11) and of the high-T_c oxides, do not play an important role in the relaxation effects described here.

In conclusion, the dc magnetization in $CeCu_2Si_2$ is unstable at $H<H_{c1}$. After a field H_i is applied in a virgin specimen, bulk or powdered, the magnetization relaxes clearly following the law $M-M(t_0) \propto H_i^n \ln(t/t_0)$ with $n=4$. If the field H_i is removed, the same law is followed at $H=0$ with a logarithmic rate $\partial M/\partial \ln t$ about a factor of 3 smaller. The origin of such behavior remains to be solved.

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