The anomalous mixed state of the C15-Laves phase superconductor $CeRu_2$: II. History dependence in field-cooled magnetization hysteresis

1998 J. Phys.: Condens. Matter 10 8327

(http://iopscience.iop.org/0953-8984/10/37/018)

J. Phys.: Condens. Matter **10** (1998) 8327–8340. Printed in the UK PII: S0953-8984(98)93550-0

The anomalous mixed state of the C15–Laves phase superconductor $CeRu_2$: II. History dependence in field-cooled magnetization hysteresis

S B Roy, P Chaddah and Sujeet Chaudhary

Low Temperature Physics Group, Centre for Advanced Technology, Indore 452013, India

Received 22 April 1998, in final form 17 June 1998

Abstract. We report an unusual history dependence of the field-cooled magnetization in and around the peak-effect regime of the superconducting mixed state of $CeRu_2$. Such history effects in magnetization are shown to be absent in the peak-effect regime of the high-T_c superconductor Bi-2212. Some qualitative similarities with and at the same time considerable quantitative differences from relatively old studies of the history dependence of the transport critical current density (J_c) in single-crystal Nb and Nb_3Ge and Mo_3Si amorphous films are examined. Our work indicates that the peak effect in $CeRu_2$ is probably due to a first-order phase transition unlike the peak effect in Bi-2212, and that the latter could be due to a continuous or second-order phase transition. Differences in details of the history effects suggest that the origin of the peak effect in $CeRu_2$ is likely to be different from those for these hard type-II superconductors.

1. Introduction

In a recent paper [1] henceforth termed I, we presented a detailed magnetization study addressing various questions regarding the anomalous (H, T) or peak-effect (PE) regime of the C15–Laves phase superconductor $CeRu_2$. We showed that a picture of dynamic crossover of pinning properties alone was inadequate to explain the anomalous behaviour of $CeRu_2$, and a thermodynamic first-order phase transition could explain the observed behaviour in a better way. The debate as to whether a PE can arise due to a thermodynamic phase transition in the vortex matter has become very interesting with the recent suggestion of such phase transitions in the PE region of various high-T_c superconductors (HTSC)—Bi-2212 [2], Y-123 [3] and Nd–Ce–Cu–O [4]. In the recent papers, the PE is believed to be associated with a second-order phase transition from an ordered (quasilattice or Bragg glass) to a disordered vortex lattice. It should be recalled here that until at least 1995, various kinds of dynamical crossover in the pinning properties [5–8] were thought to be responsible for the PE in HTSC materials.

In this paper we present an unusual history dependence of field-cooled (FC) magnetization in and around the PE regime of $CeRu_2$, which (to our knowledge) has not been reported for any other hard type-II superconductor, including the recently well studied HTSC samples. These results, we believe, will strengthen various arguments presented in I and indicate the possibility of a newer kind of vortex matter phase transition with interesting history effects in the mixed state of type-II superconductors.

The rest of the paper is organized in the following way. In section 2 we present details of the $CeRu_2$ samples used in the present study, and of our dc magnetization measurement. In section 3 we present a detailed study of the FC magnetization M_{FC} and minor hysteresis loops (MHL) obtained from the FC mode. Then we present the results of the same study for a single-crystal sample of Bi-2212, which will help to highlight the unusual features observed for $CeRu_2$. At this stage we recall that the PE has been observed in many conventional superconductors [9], and one possible origin of the PE is a softening of the Abrikosov flux lattice [10, 11], with various possible causes for this softening [12]. Similarly, it has been recognized that the PEs in $CeRu_2$ and in HTSC could also have different origins [13]. While some qualitative differences between $CeRu_2$ and HTSC have been brought out in I, some more will be brought out in section 3. We will then discuss in detail two relatively old reports on the effect of field history on the transport critical current J_c in single-crystal samples of Nb [14] and in amorphous films of Nb_3Ge and Mo_3Si [12] in the context of our present results. We shall conclude by combining the results of our previous study I and section 3 as a basis for commenting on the nature of PE in $CeRu_2$.

2. Experimental procedure

In the present study, we have used two polycrystalline samples of $CeRu_2$ (with T_c in the range 6.0–6.1 K) obtained from different sources (Los Alamos National Laboratory and the University of Kentucky), one $(Ce_{0.95}Nd_{0.05})Ru_2$ sample ($T_c \approx 6.8$ K) obtained from Imperial College, London, and a single-crystal sample of Bi-2212 ($T_c \approx 89$ K) obtained from the University of Warwick. Details of the sample preparation, characterization, and the justification of the usage of these samples in the study of PE in $CeRu_2$ can be found in I.

The DC magnetization measurements were performed with a commercial SQUID magnetometer (Quantum Design MPMS-5) using a single scan of 2 cm length in the 'fixed-range' mode. The field inhomogeneity 'seen' by the sample in a 2 cm scan is ≈2 Oe when the applied field is 20 kOe. The sample is thus forced to scan a minor hysteresis loop as it 'sees' this 2 Oe field variation during the measurement process [1]. The slight variation in magnetization during the measurement scan produces a systematic error in the inferred magnetization, but this error is negligible as long as the field for full penetration is much larger than 2 Oe [1].

Before the start of each experimental cycle, the sample chamber was heated to above 150 K and flushed with helium gas. This was done in order to minimize the effect of the suspected small amount of oxygen leaking into the sample chamber over a period of time. This operation is particularly important for the pure $CeRu_2$ samples, for which the M–H curves for a substantial field regime (which includes the PE regime of present interest) remain very near to the $M = 0$ baseline. Unless the sample chamber is flushed with helium at an elevated temperature, the magnetic contribution from the oxygen accumulated over a period of 24 hours can cause a change in the magnitude of M in this field regime by 10% on the scale of 2.5×10^{-3} emu. (This change, however, does not seem to affect the present results qualitatively.) For the 5% Nd-doped $CeRu_2$ sample, the M–H curves in the field regime of present interest lie well away from the $M = 0$ baseline. For this sample, with a relatively large value of the magnetization, the effect of oxygen is found to be virtually negligible; however, as good practice, here we also followed the regular routine of flushing the sample chamber with helium at an elevated temperature. Some further relevant details of the magnetization measurements can be found in I.

3. Results and discussion

3.1. Comparative study of the FC magnetization and minor hysteresis loops in $CeRu_2$ and Bi-2212

We have shown earlier (see I and the references therein) that, at the onset of the PE regime of $CeRu_2$, the behaviour of the minor hysteresis loops MHL_{ZFC} initiated from various values of the field (H) on the ascending-field (or lower) envelope curve (obtained after zero-field cooling (ZFC)) does not conform with the critical-state models (CSM); that is, they saturate without touching the descending-field (or upper) envelope curve. We recall here that, within the CSM, the magnetization in a MHL can only reach saturation when the shielding currents flow in the same sense everywhere. MHLs which start on the lower (upper) envelope curve will saturate when the field is lowered (raised) by the field for full penetration (H_{II}). The saturation hysteresis ΔM_H reflects the pinning properties and depends on the product of the critical current (J_c) and the sample size (D). The MHL initiated well inside the PE regime at fields larger than H_P (where H_P is the field at which the hysteretic magnetization or the inferred J_c reaches its maximum value) is in accord with the CSM, and touches the upper envelope curve when the field excursion is beyond H_{II}. The normal behaviour is of course also observed in the usual irreversible (H, T) regime.

We shall now study the magnetization (M) and MHLs within the following experimental protocols.

(i) The field, at which the measurement has to be made, is switched on and stabilized at $T > T_c$.

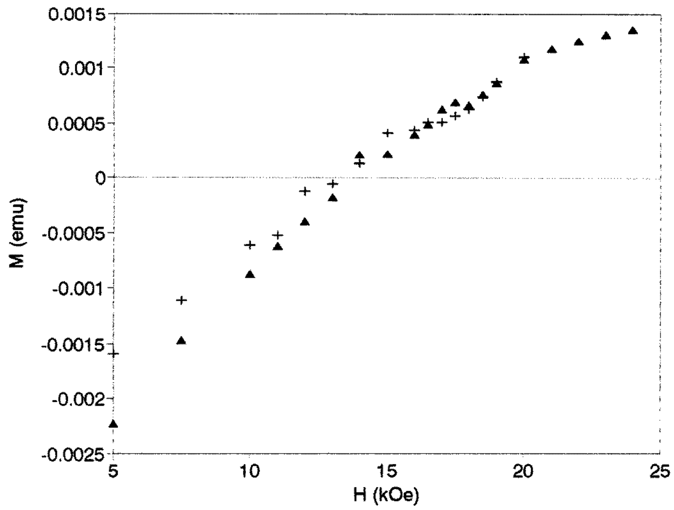
(ii) With this field on, the temperature is lowered to the temperature of measurement. This is the so-called FC mode.

(iii) After the required (H, T) point in the FC mode has been reached, an MHL is generated either by raising H ($MHL_{FC\uparrow}$) or by lowering H ($MHL_{FC\downarrow}$) isothermally.

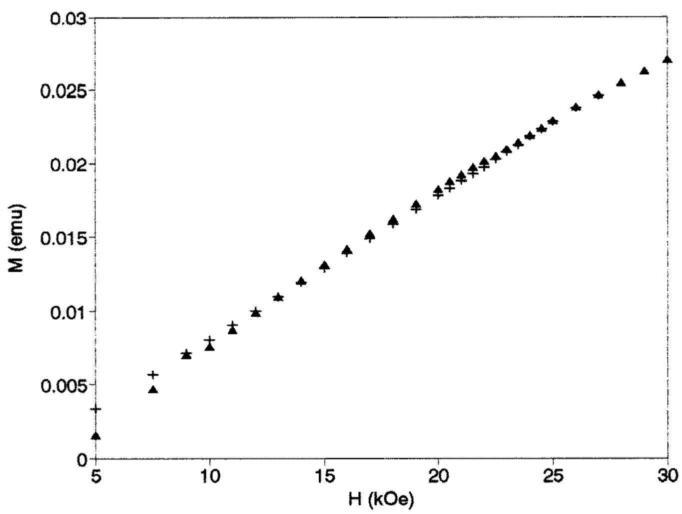
The M_{FC}s and MHL_{FC}s thus obtained will be compared with the isothermal ZFC magnetization (M_{ZFC}) and MHL_{ZFC} for various (H, T) regimes for $CeRu_2$.

The isothermal M–H curve is obtained by raising the field from below $-H_{C2}$ to 0 to above H_{C2} (the lower envelope curve), and then lowering it to 0 (the upper envelope curve). According to conventional wisdom, the set of mid-points of the envelope curve give the equilibrium M–H curve at a particular T. In the case of M_{FC}, because of flux trapping during field cooling, one actually obtains a value which is above the mid-point of the envelope curve (at the corresponding field). The MHL_{FC} generated from such FC points reaches the envelope curves on varying the field by H_{II}. These are predictions of the CSM, and are actually observed for various hard type-II superconductors including Nb and HTSC materials [15]. Our measurements in the usual irreversible regime of the present $CeRu_2$ samples and the 5% Nd-doped $CeRu_2$ sample are consistent with such predictions of the CSM, and indicate that the pinning-related physical properties in the regime concerned are those of a conventional hard type-II superconductor.

A marked deviation from the normal behaviour of M_{FC} is, however, observed for the $CeRu_2$ samples over a substantial field regime, which starts above the usual irreversible regime and is extended to well inside the PE regime. In this field regime, M_{FC} lies distinctly below the mid-point of the envelope curve. To show this anomalous effect, in figure 1 we plot the mid-point of the envelope curve, and M_{FC} for the $CeRu_2$ sample from Los Alamos (henceforth this will be termed MD1) and the $(Ce_{0.95}Nd_{0.05})Ru_2$ sample from Imperial College (henceforth this will be termed ICND) for the same reduced temperature $T/T_c \approx 0.735$. ($T = 4.5$ K (5 K) and $T_c \approx 6.1$ K (6.8 K) for the sample MD1 (ICND).)



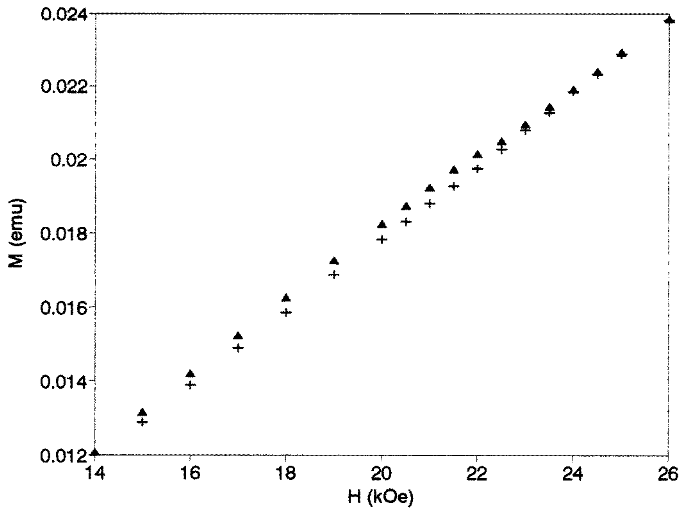
(a)



(b)

Figure 1. The mid-point of the envelope curve (filled triangles) and the field-cooled magnetization (+) versus field plots for (a) the sample MD1 at $T = 4.5$ K and (b) the sample ICND at $T = 5$ K (see the text for details). (c) shows the anomalous regime of the ICND sample on an expanded scale. The masses of the samples MD1 and ICND are 0.0295 and 0.0503 g respectively. The magnetization presented in these as well as the other figures is for the (respective) total sample mass.

This anomalous aspect of M_{FC} for the ICND sample at $T = 4.5$ K has been reported earlier [16]. We have measured the time dependence of this anomalous M_{FC} for the samples MD1 (at $T = 4.5$ K and $H = 17$ kOe) and ICND (at $T = 5$ K and $H = 21$ kOe); it shows a roughly logarithmic increase with time (see figure 2). Since the time-dependent changes in M are expected to be small, we shall briefly discuss the errors introduced by the sample's excursion, in a slightly inhomogeneous field, during the measurement process.



(c)

Figure 1. (Continued)

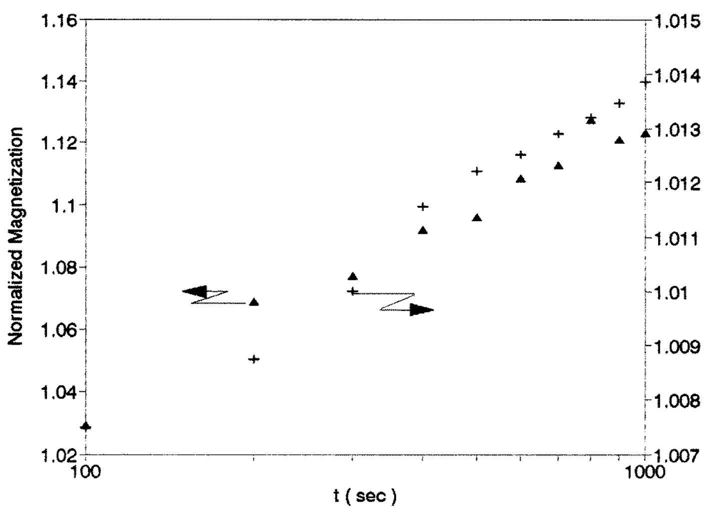
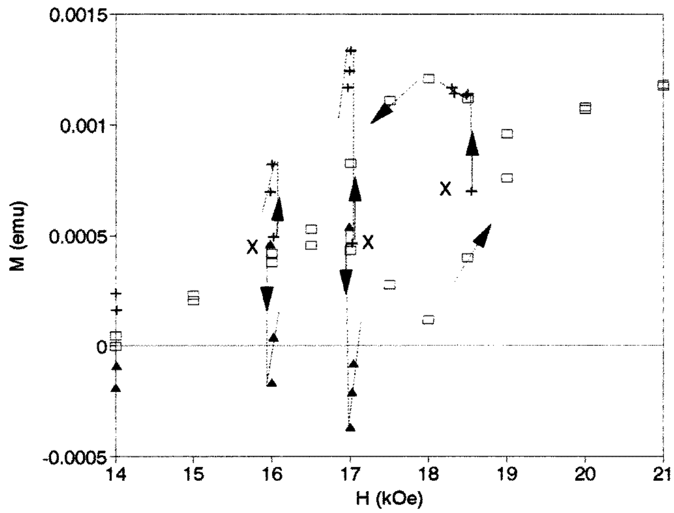
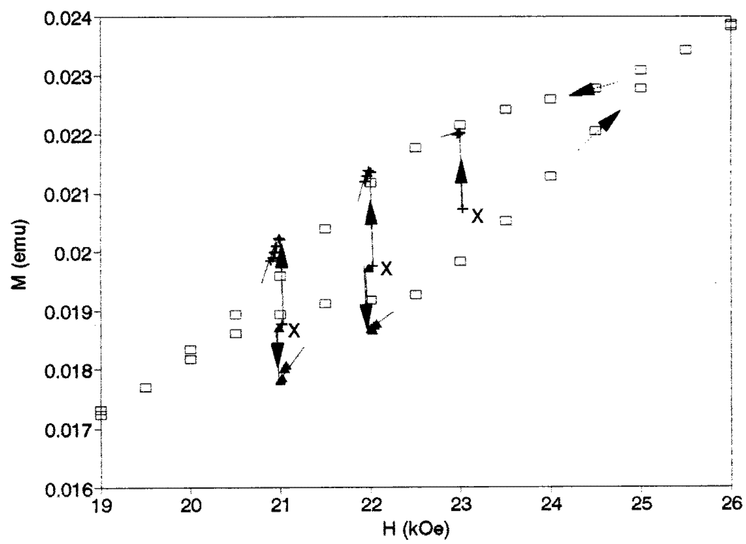


Figure 2. The normalized field-cooled magnetization versus time plot for the samples MD1 (at $T = 4.5$ K and $H = 17$ kOe) (filled triangles) and ICND (at $T = 5$ K and $H = 21$ kOe) (+).

We first emphasize that if there was no time-dependent decay of the shielding currents, then the measured response would be identical in the second and subsequent measurements irrespective of the field inhomogeneity that the sample experiences during the measurement scan. The error introduced by the field inhomogeneity is systematic and not random. The time decay of the shielding currents, however, does introduce a small relative error between successive measurements. The field inhomogeneity of about 2 Oe in our case sets up fresh shielding currents in the surface region of the sample during every measurement, and the contribution to the magnetization from the shielding currents in this surface region does

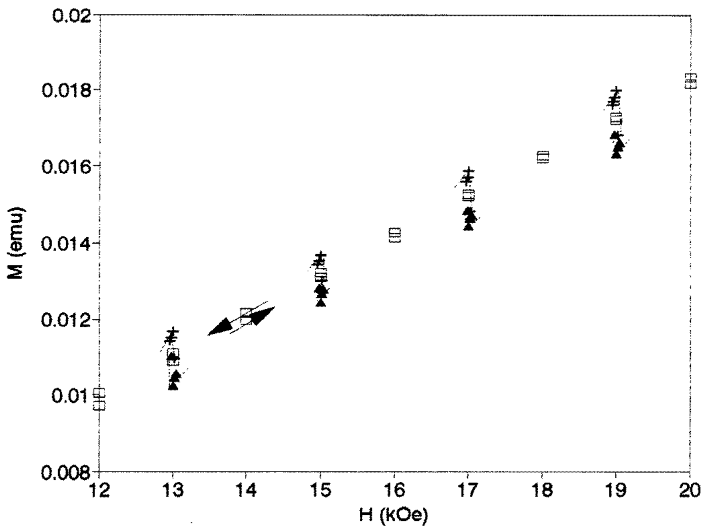


(a)



(b)

Figure 3. (a) The forward legs of some representative minor hysteresis loops, $MHL_{FC\downarrow}$ (represented by $+$) initiated at $H = 14.02$, 16.02, 17.02, and 18.5 kOe and $MHL_{FC\uparrow}$ (represented by filled triangles) initiated at $H = 13.98$, 15.98, and 16.98 kOe, for the sample MD1 at $T = 4.5$ K. The starting point of each MHL is indicated as X. Squares represent the envelope curve. Since MHLs are taken by varying the field in steps of 20 Oe, some dashed lines with arrows are provided as a guide to the eye. (b) The forward legs of some representative minor hysteresis loops, $MHL_{FC\downarrow}$ (represented by $+$) initiated at $H = 21.02$, 22.02, and 23.02 kOe and $MHL_{FC\uparrow}$ (represented by filled triangles) initiated at $H = 20.98$ and 21.98 kOe, for the sample ICND at $T = 5$ K. The starting point of each MHL is indicated as X. Squares represent the envelope curve. (c) Some more MHL_{FC}s for the sample ICND at $T = 5$ K in the field regime for which the zero-field-cooled isothermal magnetization shows almost reversible behaviour. The respective symbols for the different MHL_{FC}s and the envelope curve are the same as in (b).



(c)

Figure 3. (Continued)

not appear to decay. The observed temporal change in magnetization is thus (slightly) less than the actual change that would have been seen if the field inhomogeneity were zero. The other possible source of error is a slight temperature variation (<0.02 K) over the period of the decay time, and this may cause some random error. The qualitative feature of M_{FC} increasing with time is thus unaffected by the slight inhomogeneous field of the magnet. This change in M_{FC} is again in contrast with what has been observed for the usual irreversible regime of the present samples and also for other hard type-II superconductors, where a distinct decrease of M_{FC} with time is actually observed. All of these anomalous behaviours cease to occur well inside the PE regime at fields larger than the field H_P. It is interesting to note that M_{FC} in this higher-field regime almost matches with the mid-point of the envelope curve, indicating almost complete Meissner-flux expulsion. This is in contrast with the usual irreversible regime with comparable magnitude of magnetization irreversibility, for which M_{FC} lies distinctly above the equilibrium magnetization, indicating incomplete Meissner-flux expulsion.

The most important observation, however, is the distinctly anomalous character of the MHL$_{FC}$s in the field regime mentioned above. In figure 3 we plot MHL$_{FC\downarrow}$ and MHL$_{FC\uparrow}$ initiated from various field-cooled points for the samples MD1 and ICND. The MHL$_{FC\downarrow}$s and MHL$_{FC\uparrow}$s initiated at the lower-field end of the PE regime extend well beyond the upper and the lower envelope curves respectively. (The observed behaviour does not depend on the rate of cooling to reach the desired temperature from above $T_c(H)$.) This is clearly in contradiction with what is expected within the CSM. In the higher-field regime of the PE (i.e. for $H > H_P$) and in the usual irreversible regime, the MHL$_{FC}$s are in accord with the CSM; that is, they saturate by reaching the respective envelope curves. Qualitatively similar results have been observed for the sample (KY1) obtained from the University of Kentucky. The most unusual result, however, is the clear presence of relatively large irreversibility in MHL$_{FC}$ in the field regime well below the PE regime (see figure 3). In a substantial part of this field regime, the isothermal ZFC magnetization shows relatively low (in the sample MD1) or almost zero (in the sample ICND) irreversibility.

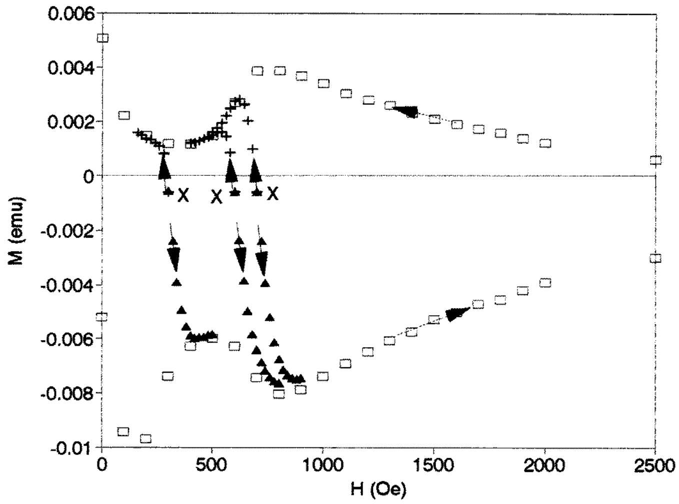


Figure 4. The M–H hysteresis curve (□) of Bi-2212 single crystal taken at $T = 25$ K with $\boldsymbol{H} \parallel c$-axis. We show forward legs of $MHL_{FC\downarrow}$ (+) and $MHL_{FC\uparrow}$ (filled triangles) initiated at the field-cooled points (marked X) at 300, 600, and 700 Oe. Note that all of these MHLs merge with the envelope curve.

These anomalous properties of M_{FC} and MHL_{FC} have not, to our knowledge, been observed so far for any other hard type-II superconductor showing a PE. To reinforce this point, in figure 4 we show MHL_{FC}s in and around the PE regime of Bi-2212 single crystal. (In our earlier paper I, we showed the MHL_{ZFC}s obtained for the same sample.) It is quite clear in figure 4 that all of the MHL_{FC}s are in accord with the predictions of the CSM; they saturate by reaching the envelope curve. It is to be noted here also that, unlike for $CeRu_2$, M_{FC} always lies above the mean value of the envelope curve for Bi-2212, as expected due to the incomplete Meissner-flux expulsion.

3.2. Dynamical crossover in pinning properties or a thermodynamic phase transition?

While obtaining the results in our previous study I and in the present study, we prepared the flux-line lattice within the following distinct experimental protocols.

(i) Zero-field cool the sample, switch on a field less than $-H_{C2}$, and then increase the field isothermally to reach various points on the lower envelope curve.

(ii) After the above step, increase the field to a value greater than H_{C2}, and then reduce the field to various points on the upper envelope curve, while always maintaining the isothermal condition.

(iii) Field cool the sample in various (positive) fields from a temperature substantially above T_c, and then lower (raise) the field in steps to reach the upper (lower) envelope curve.

We assert here that, within these experimental protocols, the observed field–temperature history effects are reproducible (within the error bar) over many experimental cycles. A dynamic transition cannot explain these definite field–temperature history effects in the MHLs in any straightforward manner, unless one invokes the idea of a field–temperature dependence of the critical current density (J_c). This in turn implies that there exist history effects of the pinning property itself. It should be noted here that such a history dependence of pinning properties is not a necessary consequence of a dynamic transition. There exists, however, a very interesting experimental study of history effects on the transport $J_c(H)$ in a single crystal of Nb by Steingart *et al* [14], which certainly deserves serious attention in the context of our present study. Following very similar experimental protocols (to those in the present case), it was clearly shown that $J_c(H)$ was larger when the sample was heated above $T_c(H)$ and then cooled to the desired temperature in a constant magnetic field. Steingart *et al* [14] argued as follows: '*Since the shear modulus of the flux-line lattice (FLL) is very small near H_{C2}, instead of a 'single-crystal' FLL being formed in the presence of pins, it is likely that the FLL will form in grain structure where the local orientation of the FLL is such that the number of net pins in flux-line cores is maximized in each grain. In this way an increase in the negative interaction energy between flux lines and pins will be achieved at the expense of a small addition of FLL defect energy*.' This special granular structure which forms at T infinitesimally smaller than $T_c(H)$ has the largest number of effective pins, and hence the largest J_c. Changes in the FLL parameter within a particular FLL grain after formation will cause a local decrease in the number of effective pins. Within the above hypothesis, Steingart *et al* [14] tried to explain the enhancement of $J_c(H)$ for the different methods of FLL formation. They also observed the disappearance of the history effects in the higher-field regime of the PE at H_P, where $J_c(H)$ and the pinning force F_P reached their peak values. This behaviour was explained in terms of the flux-pinning model of Kramer [17], in which the FLL shear around the strongest line pins rather than breaking them at fields greater than H_P. In Kramer's model the dynamic pinning force above H_P depends strongly on the magnitude of C_{66} and is largely insensitive to the density of line pins. Generation of an increased number of effective pins by field cooling is not expected to increase F_P in this field regime. This picture for Nb at first glance seems to be very appropriate for explaining the unusual field–temperature history effects in $CeRu_2$ also. On closer inspection, however, there are seen to exist some distinct features in $CeRu_2$ which will discourage acceptance of this picture as such; these we shall now elaborate on.

To elaborate on the history effects in Nb, Steingart *et al* [14] introduced an enhancement factor

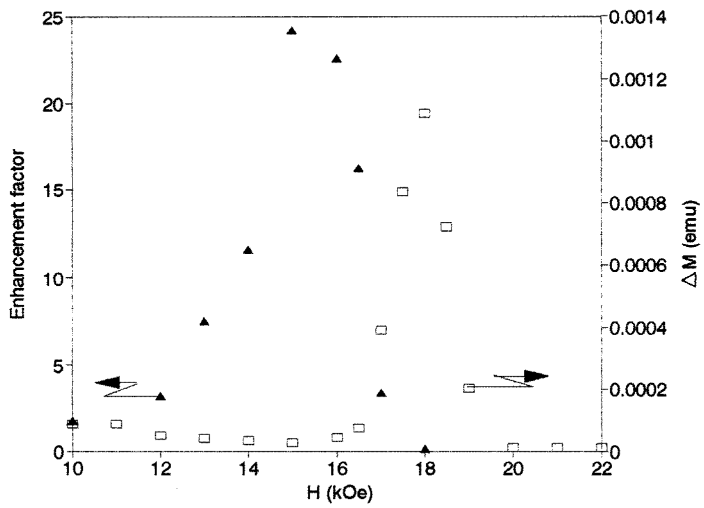
$$\epsilon = [J_c(\mathrm{FC}) - J_c(\mathrm{FF})]/J_c(\mathrm{FF})$$

where J_c(FC) is the critical current measured after the FLL has formed by field cooling and J_c(FF) is the critical current measured by extrapolation of the flux-flow portion of the V–I curve to $V = 0.75\ \mu$V. When plotted as a function of H, the enhancement factor shows a substantial value and a peak in the region where $J_c(H)$ shows a PE, and is small elsewhere. We shall define a similar enhancement factor

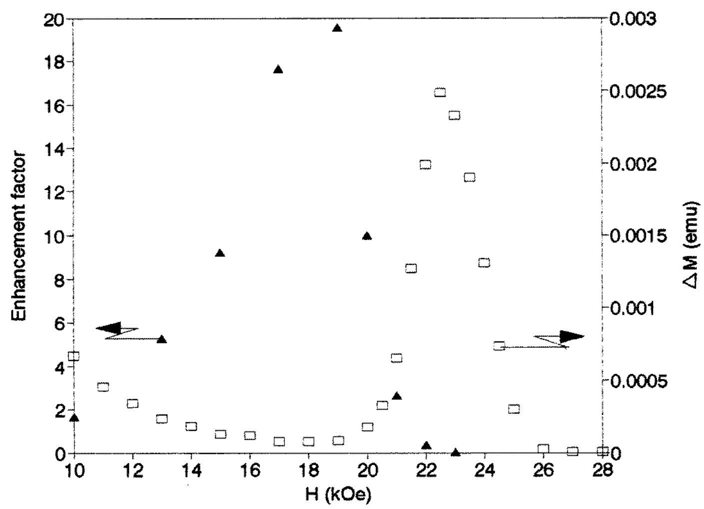
$$\epsilon = [\Delta M_H(\mathrm{FC}) - \Delta M_H(\mathrm{ZFC})]/\Delta M_H(\mathrm{ZFC})$$

for our $CeRu_2$ samples, substituting for the critical current the saturation magnetization hysteresis value (ΔM_H) of the various MHLs, assuming implicitly that D shows no history effects. However, note that there is no counterpart of J_c(FF) in the magnetization hysteresis; we have taken instead ΔM_H(ZFC), which is the counterpart of the critical current J_c measured after the FLL has been prepared by zero-field cooling and subsequent isothermal variation of the field. The inferred J_c obtained in such a way is greater than J_c(FF); hence the estimated enhancement factor will be somewhat underestimated. When such an enhancement factor is plotted for the samples MD1 and ICND, it shows a very large value in a field region quite far away from the PE regime (see figure 5).

Similar history effects in the transport J_c in and around the PE regime have also been reported for amorphous films of Nb_3Ge and Mo_3Si [12]. It was argued that the presence



(a)



(b)

Figure 5. The enhancement factor (ϵ) (see the text for details) versus magnetization (H) plot (a) for the sample MD1 at 4.5 K and (b) for the sample ICND at $T = 5$ K. Also shown in the figures is the field dependence of the magnetization hysteresis (ΔM_H) of the zero-field-cooled isothermal magnetization, displaying a peak effect at around 18 kOe (22.5 kOe) for the sample MD1 (ICND). Note that ϵ peaks well below the peak in ΔM_H.

of flux-line dislocations in the PE regime caused an enhancement of the pinning force by the local reduction of the shear modulus. Here, according to Wordenweber *et al* [12] '*a structural transition from mainly elastically to a plastically distorted lattice occurs*'. It was shown that around the PE regime a flux-line lattice could exist in metastable states, leading to a history dependence of the pinning force and J_c. To compare with these results, in figure 6 we plot the volume pinning force F_P (estimated from the M–H data as $F_P = J_c \mu H \propto \Delta M_S H$) as a function of the reduced field ($h = H/H_{C2}$) for our samples

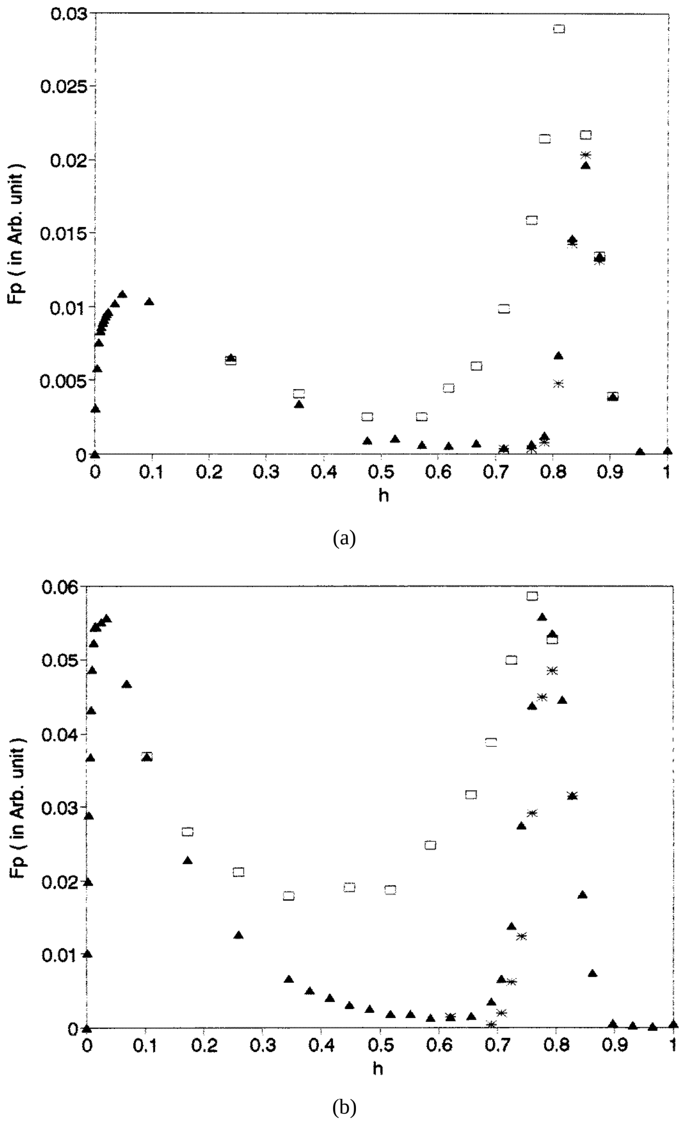


Figure 6. The pinning force (F_P) versus reduced field (h) plots for the samples (a) MD1 and (b) ICND. Asterisks stand for F_P obtained in the ascending-field cycle after ZFC, filled triangles stand for F_P obtained in the descending-field cycle after ZFC and an excursion to a field greater than H_{C2}, and squares stand for F_P obtained in the FC mode.

MD1 and ICND. ΔM_s is estimated for each H from the difference between the saturation magnetizations of the minor hysteresis loops centred at that H. These MHLs ($MHL_{ZFC\downarrow}$s, $MHL_{ZFC\uparrow}$s, and MHL_{FC}s) are obtained after preparing the FLL under three different sets of conditions following the experimental protocols described at the beginning of section 3.2. In the field regime where no history effects are observed, the MHLs obtained under the three different experimental protocols saturate on merging with the envelope curve; in this situation ΔM_S stands for the difference between the upper and lower envelope curve. The differences in the history effect between $CeRu_2$ and amorphous Nb_3Ge films are listed below.

(i) The history effect in the ZFC isothermal field cycle is confined to a relatively narrow field regime for our present samples (roughly between 16 and 18 kOe for MD1 and 20 and 24 kOe for ICND) between the onset of the PE regime and H_P. This is to be contrasted with the case for amorphous Nb_3Ge film [12], for which a history effect in $J_c(H)$ was observed even below the PE regime (see figure 6 of reference [12]).

(ii) There exists a major difference in the shape of $F_P(h)$ itself between these two cases. While the $F_P(h)$ curve for amorphous Nb_3Ge films shows a single-peak structure which coincides with the peak in $J_c(h)$, a distinct two-peak structure with very low values between the peaks is observed in $F_P(h)$ for many samples of $CeRu_2$ showing a PE [18], including the present one (see figure 6).

(iii) We see a large difference in the values of $F_P(h)$ obtained in the FC mode, which starts immediately below H_P and persists well outside the PE regime. No study of $F_P(h)$ obtained in the FC mode has been reported for the amorphous Nb_3Ge films.

With this information, it is not very apparent that the picture of metastable FLL and the associated dynamics proposed for these amorphous films of superconductors will also be applicable to the present case of $CeRu_2$.

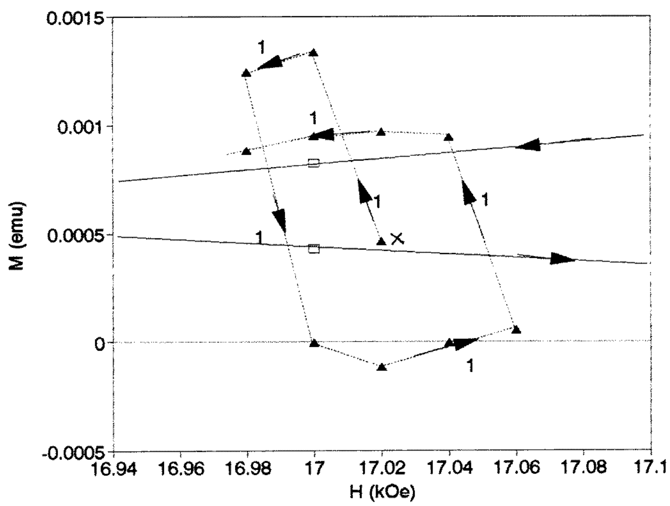


Figure 7. A MHL cycle (filled triangles) for the sample MD1, starting from the field-cooled point (indicated by X) at 17.02 kOe and following the excursion path (trace 1 marked by dashed lines) $H \rightarrow 16.98 \rightarrow 17.06$ kOe and then back to 16.98 kOe in 0.02 kOe steps (see the text for details). The envelope curve is represented by the solid lines through the points □.

Let us now consider the alternative approach of using a first-order phase transition [19, 20] to explain the observed anomalous behaviour of MHL_{FC}. We recall here that in our previous study I, we argued that in the ZFC isothermal magnetization cycle the observed magnetic response at the onset of the PE regime supports the idea of a first-order thermodynamic transition into a new superconducting state with enhanced pinning. This new superconducting state is nucleated in domains whose physical size grows on increasing the field, and which supercool on decreasing the field. At this stage we emphasize that we are only considering a first-order phase transition and not the exact microscopic structure of the superconducting state as a result of this transition. In our picture any superconducting state with enhanced pinning is equally valid, irrespective of whether this enhanced pinning is due to a new kind of superconducting order parameter [19, 20], due to super-softening of the FLL [21], due to a vortex phase transition [2–4], or due to any other cause hitherto unknown. Our new MHL_{FC} results fit into this picture in a relatively smooth manner. In the field-cooled mode the higher-field–temperature phase supercools and is carried into the (H, T) regime far beyond the PE regime (obtained with a ZFC isothermal field cycle). There exist indications that this supercooled phase shatters, on cycling the external field. To show this effect, in figure 7 we plot a complete cycle of $MHL_{FC\downarrow}$ for the MD1 sample initiated at $H = 17.02$ kOe and followed by the field excursion $H \rightarrow 16.98 \rightarrow 17.06 \rightarrow 16.98$ kOe. It is clear that the saturation magnetization obtained at 17 kOe in the forward leg of the MHL is larger than that obtained when the MHL reaches 17 kOe in the process of completing the full cycle (following trace 1 in figure 7). For the sake of clarity and conciseness, we have presented only one shattered cycle for the sample MD1, although there are qualitatively similar results for other fields and for the other samples also. These results suggest that the MHL_{FC}s shatter quite easily on cycling the applied field. We have shown earlier, in I, the similar shattering of the MHL_{ZFC}s initiated from the upper envelope curve.

It should be noted here that, as far as the enhanced pinning is concerned, the picture of a granular FLL in the field-cooled mode of Steingart et al [14] is equally valid for our high-field–temperature superconducting phase. The additional idea of a first-order transition and an associated supercooling phenomenon just provides a natural explanation of the field-cooled MHL_{FC}s in the field regime well away from the PE regime and the shattering of these MHL_{FC}s on field cycling. Here we would like to add some words of caution regarding the experimental study of a first-order phase transition. To monitor a first-order transition it is required that the external parameter (in the present case the magnetic field), which actually induces the transition, should not be cycled rapidly while traversing the transition regime. An ac susceptibility measurement after preparing the FLL at the desired (H, T) point does not seem to fulfil such a condition, since the measurement involves a rapid field cycling. A fair amount of caution is necessary in the transport measurements as well. Since the transport current induces a magnetic field, this current will be another external parameter in preparing the FLL at the desired (H, T) point, and one has to be careful to make a comparison with the dc magnetization measurement. Hence, as far as the question of a first-order transition is concerned, and for a real comparison with the history effects in Nb, it is desirable to have a dc magnetization measurement performed on Nb samples as well as amorphous Nb_3Ge and Mo_3Si films. In our dc magnetization measurement on an Fe-doped polycrystalline Nb sample, however, we failed to observe any unusual history effects (see also I). A detailed measurement on a single-crystal sample of Nb (showing a PE) is now called for.

4. Conclusion

Summarizing our results, we can say that we have seen unusual history effects in $CeRu_2$ in the field-cooled magnetization and minor hysteresis loops initiated from these field-cooled points, which start in a field regime well below the PE regime and continue until $H < H_P$ in the PE regime. In I we had shown history effects in the ZFC case, but restricted to $H < H_P$ within the PE regime. Both of these types of history effect have been shown to be absent in the PE regime of Bi-2212, and we can conclude that such history effects are not an essential consequence of the PE. Some qualitative similarities to (but with differences in detail from) relatively old studies of J_c in single crystals of Nb [14], and amorphous Nb_3Ge and Mo_3Si films [12], have been examined. Our work indicates that the PE in $CeRu_2$ is probably due to a first-order phase transition unlike the PE in Bi-2212, and that the latter could be due to a continuous or second-order phase transition. The history effects in J_c

seen earlier [12, 14] might imply that the peak effects in some conventional superconductors could also be accompanied by a first-order phase transition, and magnetization studies of these superconductors are called for. Differences in detail of the history effects imply that the origin of the PE in $CeRu_2$ may be different from that of the Nb, and the possible causes discussed in I need to be investigated.

Acknowledgments

We thank Professor J W Lynn and Professor L E DeLong for providing us with well characterized samples of $CeRu_2$. Materials support provided by late Professor B R Coles and many useful discussions with him are gratefully acknowledged. We also thank Dr G Balakrishnan for providing us with the single-crystal sample of Bi-2212.

Note added in proof. A recent paper (Ravikumar G *et al* 1998 *Phys. Rev.* B **57** R11 069) shows anomalies in the FC magnetization curve as the field is reduced, in the PE regime of $CeRu_2$ and $NbSe_2$.

References

[1] Roy S B, Chaddah P and Chaudhary S 1998 *J. Phys.: Condens. Matter* **10** 4885
[2] Khaykovich B, Zeldov E, Majer D, Li T W, Kes P H and Konczykowski M 1996 *Phys. Rev. Lett.* **76** 2555
[3] Deligiannis K, de Groot P A J, Oussena M, Pinfold S, Langan R, Gagnon R and Taillefer L 1997 *Phys. Rev. Lett.* **79** 2121
[4] Giller D *et al* 1997 *Phys. Rev. Lett.* **79** 2542
[5] Daumling M, Sentjeuns J M and Larbalestier D C 1990 *Nature* **346** 332
[6] Yang G, Shang P, Sutton S D, Jones I P, Abell J S and Gough C E 1993 *Phys. Rev.* B **48** 4054
[7] Tamegei T, Iye Y, Oguro I and Kishio K 1993 *Physica* C **213** 33
[8] Krusin-Elbaum L, Civale L, Vinokur V M and Holtzberg F 1992 *Phys. Rev. Lett.* **69** 2280
Yeshuran Y, Bontemps N, Burlachkov L and Kapitulnik A 1994 *Phys. Rev.* B **49** 1548
[9] Campbell A M and Evetts J E 1972 *Adv. Phys.* **21** 327
[10] Pippard A B 1969 *Phil. Mag.* **19** 217
[11] Larkin A I and Ovchinnikov Y N 1979 *J. Low Temp. Phys.* **34** 409
[12] Wordenweber R, Kes P H and Tsuei C C 1986 *Phys. Rev.* B **33** 3172
[13] Crabtree G *et al* 1997 *Phys. Essays* (submitted for the H Umezawa memorial issue)
[14] Steingart M, Putz A G and Kramer E J 1973 *J. Appl. Phys.* **44** 5580
[15] Grover A K, Paulose P L, Chaddah P and Ravikumar G 1989 *Physica* C **162–164** 337
Sarkissian B V B *et al* 1989 *Physica* C **162–164** 335
[16] Roy S B and Chaddah P 1997 *Physica* C **279** 70
[17] Kramer E J 1973 *J. Appl. Phys.* **44** 1360
[18] Chaudhary S, Roy S B and Chaddah P 1998 unpublished
[19] Modler R *et al* 1996 *Phys. Rev. Lett.* **76** 1292
[20] Steglich F *et al* 1996 *Physica* C **263** 498
[21] Carruzzo H M and Yu C 1997 *LANL Preprint* cond-mat 9705092