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2006 J. Phys.: Condens. Matter 18 9471

(http://iopscience.iop.org/0953-8984/18/41/014)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **18** (2006) 9471–9482 doi:10.1088/0953-8984/18/41/014

Temperature dependence of magnetization in the superconducting mixed state of $CeRu_2$: evidence of a first-order phase transition

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Received 2 August 2006, in final form 8 September 2006
Published 29 September 2006

Abstract
We present results of magnetization measurements in very close temperature intervals in the superconducting mixed state of $CeRu_2$. While the temperature (T) dependence of magnetization in low applied magnetic fields (H) is typical of that of a type-II superconductor with moderate pinning, the character changes markedly in high applied H, showing clear evidence of a first-order phase transition. Isothermal field-dependent magnetization studies have suggested earlier the possibility of such a first-order phase transition in the low-T high-H part of the superconducting mixed state of $CeRu_2$. The present results for the first time provide the evidence of this first-order phase transition in a temperature-dependent study of magnetization. The possible role of paramagnetic impurities in the observed phase transition is discussed.

The C15 Laves phase superconductor with a T_C of 6.2 K has drawn considerable attention during the last 50 years for various reasons. First, rare-earth (RE)-doped $CeRu_2$ alloys provided a very interesting basis for the study of the coexistence of superconductivity and magnetism [1]. Then the recognition of $CeRu_2$ as an intermediate-valence (IV) compound [2], and the related theoretical [3] studies, stimulated much interest in the normal-state properties of this compound. Further, the interesting Fermi surface topology and enhanced Pauli paramagnetism of $CeRu_2$ have given rise to various interesting possibilities, starting with an exotic non-s-wave superconducting ground state [4] to a field-induced change in the microscopic superconducting order parameter associated with the onset of a Fulde–Ferrel–Larkin–Ovchinnikov (FFLO) state [5].

In the 1990s the discovery of a striking feature in the form of an enhanced pinning of flux lines (giving rise to a large irreversibility in magnetization) in the high-field regime of the superconducting mixed state (or vortex state) of $CeRu_2$ opened up a new area of interest in the superconductivity of this compound. This striking feature was observed in various physical properties, including magnetization [6–8], magnetostriction [9], magnetotransport [10], magnetoelasticity [11] and neutron diffraction [12]. The isothermal field-induced enhancement of the irreversibility in magnetization, on first sight, closely resembles the conventional 'peak-effect' observed in various type-II superconductors. However, in a series of papers [13, 14] we have highlighted the various anomalous aspects associated with the 'peak-effect' in $CeRu_2$ which cannot be explained within a framework of pinning of the flux lines in a type-II superconductor. Further, we have shown that these anomalous features in the form of certain thermomagnetic hysteresis are actually signatures of a field-induced first-order phase transition from a vortex state with low pinning strength to a vortex state with higher pinning strength [13, 14]. Such thermomagnetic hysteresis is considered to be a good observable to characterize a disorder-influenced first-order phase transition where the discontinuity in entropy is relatively small and hence the associated latent heat is relatively difficult to measure [15]. Subsequently similar anomalous features associated with a peak-effect have also been observed in various other classes of type-II superconductor, $NbSe_2$ [16], YBaCuO [17], V_3Si [18] and MgB_2 [19], highlighting the generality of the problem.

Magnetization studies probing the 'peak-effect' in $CeRu_2$, so far, have involved mostly isothermal field variation measurements. A thermodynamic phase transition which can be induced by a magnetic field is naturally expected to be induced by temperature also. While this field-induced vortex-matter transition in $CeRu_2$ carried distinct signatures of a first-order thermodynamic phase transition, the absence of any clear signature of a temperature-induced transition has been quite intriguing. Here we present first and distinct evidence of a first-order phase transition in the superconducting mixed state of two $CeRu_2$ single-crystal samples of different purity through a temperature-dependent study of magnetization. These results will also indicate why this phase transition has evaded detection so far in temperature-dependent magnetization studies. In addition these results reveal certain interesting aspects of the superconducting mixed state of $CeRu_2$ related to the paramagnetic response of the normal state.

We have used two single-crystal samples of $CeRu_2$ in the present study. One sample with resistivity ratio of ≈15 (sample A) was obtained from Dr A D Huxley (CENG, France). The details of the sample preparation and characterization can be found in [6] and this sample has been used in our earlier measurements [14, 20, 21]. The other sample with resistivity ratio ≈210 (sample B) was obtained from Dr L E DeLong (University of Kentucky, USA). This sample actually originated from Osaka University, Japan, and the details of the sample preparation and characterization can be found in [22]. In our earlier studies of isothermal field dependence of magnetization [13, 14] we had used a SQUID magnetometer (MPMS5-Quantum Design (QD), USA). There has been quite a bit of discussion on the issue of the sample movement in the inhomogeneous field of the superconducting magnets during the process of measurement [23, 24]. In this regard we had earlier come to the conclusion that in a hard type-II superconductor, as long as the field for full penetration in the superconducting mixed state at a particular applied field is substantially greater than the field inhomogeneity encountered during the measurement, the error in the results will be negligible [13, 14, 23]. In the case of a 2 cm scan length of measurement in a QD-SQUID magnetometer, the field inhomogeneity in an applied field of 20 kOe is ≈2 Oe. This value is considerably less than the field for full penetration for $CeRu_2$ samples (say at 20 kOe), and we had used this 2 cm scan length along with other cross-checks in our earlier studies of magnetization of $CeRu_2$ [13, 14]. Even within this experimental protocol, probing the 'peak-effect' in temperature-dependent measurements using a QD-SQUID magnetometer turned out to be quite difficult [13]. We had anticipated [13] that a vibrating sample magnetometer (VSM) would be better suited for such a study, and in the present work we have now used a VSM (Quantum Design, USA). In this VSM the sample vibrates with an amplitude which can be varied from 0.2 to 4 mm and with a frequency of 20 or 40 Hz. The maximum length of sample movement in this VSM is

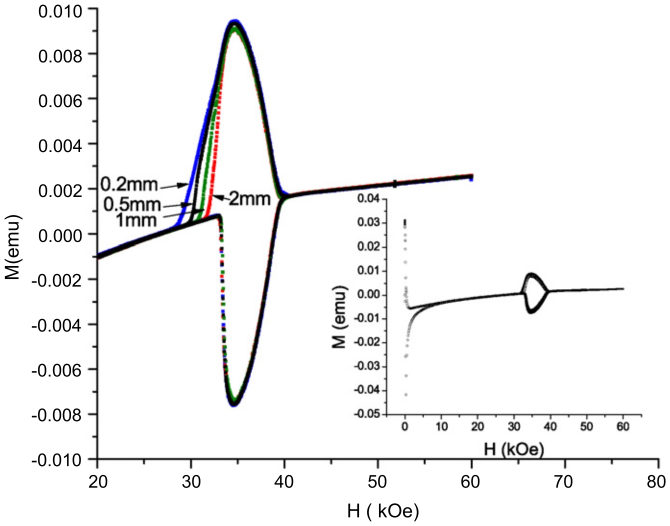

Figure 1. M versus H plot for the $CeRu_2$ single-crystal sample A at 3 K. The main figure highlights the peak-effect regime. The results are obtained with a measurement frequency of 20 Hz and with vibration amplitudes varying between 0.2 and 4 mm. The field sweep rate is 50 Oe s^{-1}. The inset shows the complete M–H curve obtained with a measuring amplitude of 1 mm and frequency 20 and 40 Hz. There is hardly any effect of variation of the measuring frequency on the M–H curve. Data presented in this figure and in the subsequent figures are actual discrete data points and not continuous guidelines.

(This figure is in colour only in the electronic version)

4 mm, and hence the field inhomogeneity encountered during the measurement procedure will be considerably smaller than that in a QD-SQUID magnetometer. Even then it is important to establish a robust experimental protocol with this new commercial VSM, and to do that we first measure the isothermal field dependence of magnetization by varying both the amplitude of vibration and the measurement frequency. In figure 1 we present the isothermal magnetization (M) versus field (H) curve at a temperature (T) = 3 K measured in sample A. First of all the typical features of superconducting response (including the peak-effect) observed earlier in this sample [6, 14] and other $CeRu_2$ samples [7, 8, 13] are reproduced here nicely. Second, there is no substantial dependence of M on the vibration amplitude, except in the H-regime where the peak-effect is turned off in the field-decreasing cycle. This latter behaviour is actually expected since, as argued earlier [13, 14], the higher-field vortex-matter phase remains as a metastable (supercooled) state in this H-regime. Any energy fluctuation will tend to convert this metastable state to an equilibrium state, and the amount of such fluctuations is likely to increase with higher amplitude. On the other hand, changing the frequency from 40 to 20 Hz did not lead to any appreciable change in the magnetization response (see the inset of figure 1). For the rest of our measurements we will use a vibration amplitude of 1 mm (the sensitivity of the VSM decreases slightly with lowering the amplitude) and vibration frequency of 20 Hz. While this experimental protocol definitely reduces the possible role of magnetic inhomogeneity and energy fluctuations on the observed non-equilibrium properties, one should still remain cautious on the possible role of energy fluctuations especially when one is dealing with an apparatus with multiple control parameters.

Before discussing the T-dependence of M it is pertinent to present the isothermal field dependence of sample B (see figure 2) and compare it with sample A. This sample has

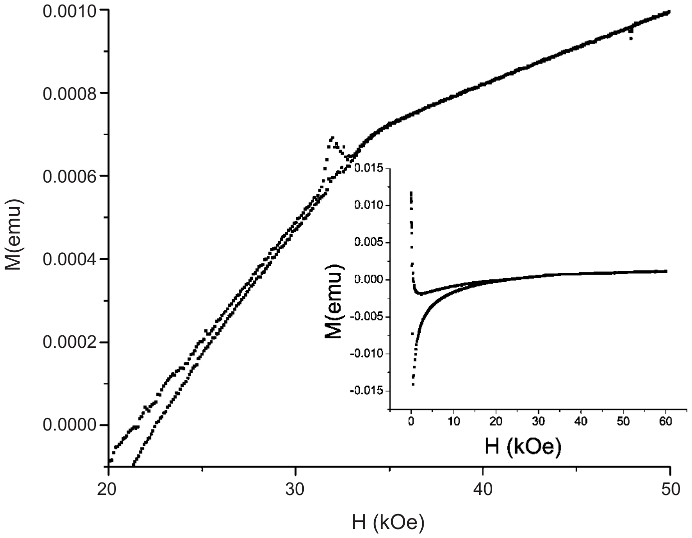

Figure 2. M versus H plot for the CeRu$_2$ single-crystal sample B at 3 K. The main figure highlights the peak-effect regime. The results are obtained with a measurement frequency of 20 Hz and with vibration amplitude 1 mm. The field sweep rate is 50 Oe s^{-1}. The inset shows the complete M–H curve.

much higher resistivity ratio and must have a much smaller amount of defects. In this sample B, however, we could not detect any peak-effect in the isothermal M–H curve when the measurement is done above 3 K. In contrast, in sample A, in various other relatively impure single crystals, and also in polycrystals of CeRu$_2$, the peak effect in a robust form is readily seen at temperatures as high as 5.5 K [6–8, 13, 14]. Also, as seen in figure 2, the peak-effect in sample B is relatively subtle even at 3 K. There exists an earlier report of such qualitatively different behaviour in very pure single crystals of CeRu$_2$ [22]. It is interesting to note that, both in that earlier work [22] as well as in the present work, irreversibility in the magnetization in the low-field regime (away from the peak-effect regime) is perceptibly higher. While the observation of more pronounced peak-effect in the less pure sample A fits well with the standard framework of flux-line pinning, the higher pinning force in the low-field regime and the relatively weak peak-effect in the high-field regime of sample B are definitely counterintuitive. We shall come back to these points later on in this paper.

Figure 3 (figure 4) presents the M versus T plots in sample A (sample B) in various applied magnetic fields (H). From low-field (100 Oe) measurements we estimate the superconducting transition temperature (T_C) to be 6.2 K (6.25 K) in sample A (sample B). In each applied field, magnetization is measured in zero-field-cooled (ZFC), field-cooled-cooling (FCC) and field-cooled-warming (FCW) mode. In the ZFC mode the sample is cooled to the lowest T of measurement (2 K in the present case) before the applied field is switched on, and the measurement is made while warming up the sample across T_C. In the FCC mode the field is switched on at a T above T_C and the measurement is made while cooling down. After reaching the lowest T the sample is warmed again keeping the field on, and this is the FCW mode. As shown in figure 3, there is a distinct difference between $M_{ZFC}(T)$, $M_{FCC}(T)$ and $M_{FCW}(T)$ below a characteristic temperature T_{irrv} when the applied field H is less than 10 kOe. Above 10 kOe, T_{irrv} in sample A is definitely below 2 K. This is a typical characteristic of type-II superconductors (in general) with pinning, and can be well explained within the framework of flux-line pinning models [25]. The same behaviour is observed in sample B. However, T_{irrv} in

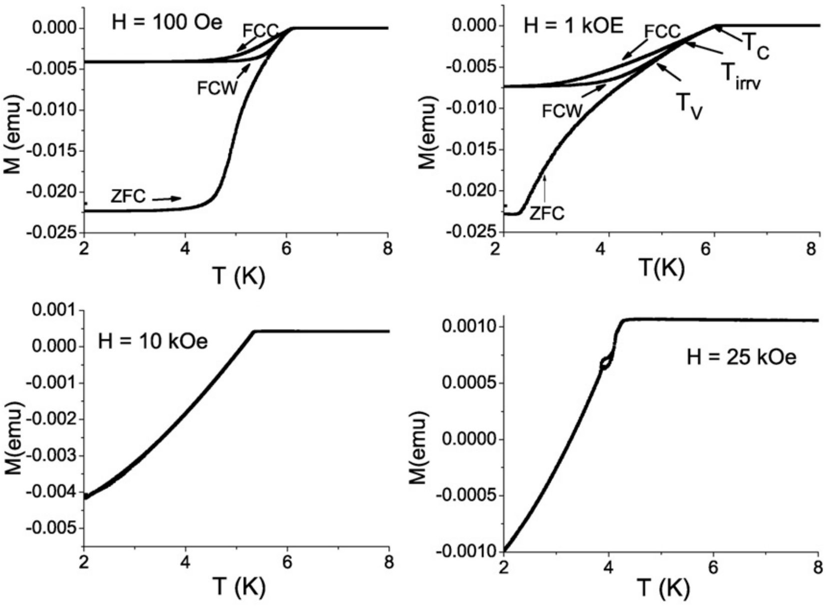

Figure 3. *M* versus *T* plots for the $CeRu_2$ single-crystal sample A in the presence of applied magnetic fields of various strength. The results are obtained with a measuring frequency of 20 Hz and vibration amplitude 1 mm. The temperature sweep rate is 0.1 K min^{-1}.

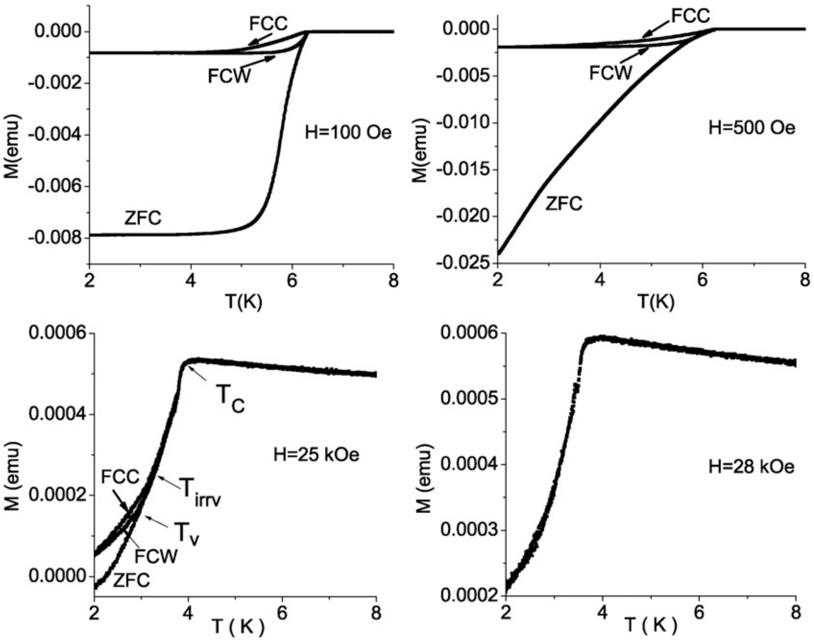

Figure 4. *M* versus *T* plots for the $CeRu_2$ single-crystal sample B in the presence of applied magnetic fields of various strength. The results are obtained with a measuring frequency of 20 Hz and vibration amplitude 1 mm. The temperature sweep rate is 0.1 K min^{-1}.

sample B remains above 2 K even in an applied field of 25 kOe (see figure 4). This observation is again counterintuitive since this sample B is supposed to have fewer defects, and hence

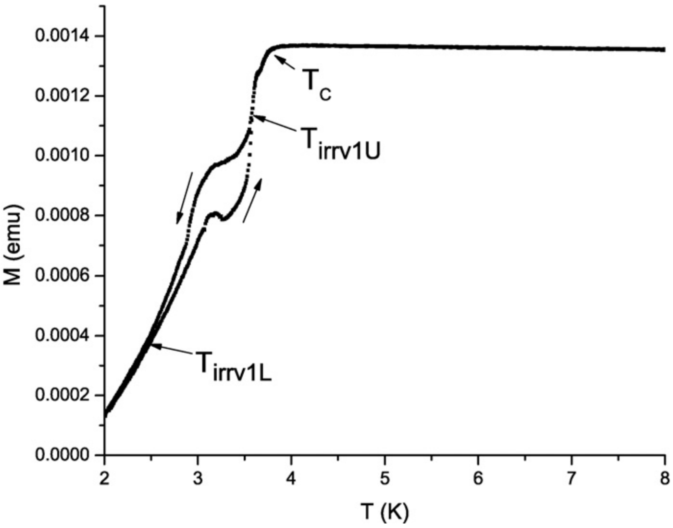

Figure 5. M versus T plot for the CeRu$_2$ single-crystal sample A in the presence of an applied magnetic field of 32 kOe, highlighting a vortex-matter phase transition and the associated thermal hysteresis. The results are obtained with a measuring frequency of 20 Hz and vibration amplitude 1 mm. The temperature sweep rate is 0.1 K min^{-1}. $T_{\mathrm{irrv}1L}(H)$ and $T_{\mathrm{irrv}1U}(H)$ represent the lower and upper limit of the temperature regime where thermal hysteresis is observed.

a smaller amount of pinning centres. Accordingly the magnetization should have been less irreversible.

As shown in figure 3, the M versus T plot for $H \geqslant 10$ kOe in sample A is independent of ZFC, FCC and FCW measurement modes, and is quite reversible in nature. However, for $H \geqslant 20$ kOe an interesting feature in the form of a hysteresis bubble appears in the otherwise reversible M–T curve of sample A (see figure 3). This feature is highlighted more in figure 5 in an M–T curve obtained with $H = 32$ kOe. In this M–T curve, with the lowering in T first there is a distinct drop in M marking the onset of the superconducting transition. This drop in M is then followed by another transition at a lower T which is marked with a distinct thermal hysteresis. In the T regime between the onset of this second transition and the onset of the superconducting transition, the magnetization is perfectly reversible (within our experimental resolution). The observed thermal hysteresis has a width of ≈ 1 K, and it is clearly seen only when the temperature variation is strictly unidirectional and with a reasonably slow (0.1 K min^{-1}) rate. Any temperature fluctuation tends to smear out the observed thermal hysteresis. A distinct reversible regime in the M–T curve appears again below this hysteresis bubble. In sample B the indication of this second transition in the M–T curve is unambiguously observed only when the applied field is 28 kOe (see figure 4). In figure 6 we present the M versus T curve for sample B in a field of 35 kOe. The second transition looks sharper in this sample, and in fact it is accompanied by a local maximum in $M(T)$. On the other hand, in comparison to sample A the associated thermal hysteresis is relatively less prominent in sample B (see the inset of figure 6).

The thermal hysteresis and the metastability[3] associated with this second transition in CeRu$_2$ clearly point toward the first-order nature of this phase transition. We recall here again that such thermal hysteresis is actually considered to be a good observable to characterize

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[3] Both sample A and sample B show large relaxation in magnetization in the temperature regime where thermal hysteresis is observed.

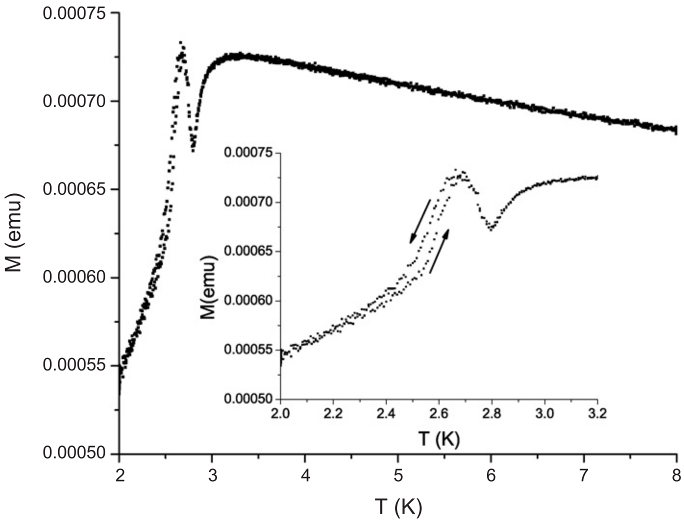

Figure 6. M versus T plot for the CeRu$_2$ single-crystal sample B in the presence of an applied magnetic field of 35 kOe, highlighting a vortex-matter phase transition and the associated thermal hysteresis. The results are obtained with a measuring frequency of 20 Hz and vibration amplitude 1 mm. The temperature sweep rate is 0.1 K min^{-1}.

a disorder-influenced first-order phase transition [15]. (And the presence of disorder in these single crystals of CeRu$_2$ has already been highlighted by the low-field irreversibility of magnetization.) Such a subtle structure (and the associated metastability) marking the phase transition and which shows up in a temperature interval of ≈1 K only, can easily evade detection unless one is carefully looking for it. Apart from having enough data points to detect the transition, both the measurement and the temperature variation process should be as little intrusive as possible so that they do not disturb the subtle aspects of this first-order phase transition process. Hence it is not surprising that this feature was not detected in earlier standard M–T measurements. We have attempted to study this transition in CeRu$_2$ with a time relaxation method of specific heat measurements. Apart from the difficulty in acquiring data points in such a close temperature interval, the heat pulse generated during the measurement procedure is actually quite intrusive in the present context. As a result we could not see any unambiguous signature of this vortex-state transition in our specific heat studies. This transition in the isothermal field-dependent magnetization study was observed only when the temperature was below a critical temperature [6–8, 13, 14]. The present temperature-dependent magnetization study, which reveals the concerned phase transition only with applied H greater than a critical value, thus confirms the earlier findings of field-dependent studies.

For a general comparison of the superconducting properties between the two single crystals of CeRu$_2$ of different purity, we plot in figure 7 the field (H)—temperature (T) phase diagram for these two samples. Here T_C represents the onset of the superconducting transition in the temperature dependence of M, and T_{irrv} is the irreversibility temperature where $M_{\mathrm{ZFC}}(T)$ (or $M_{\mathrm{FCW}}(T)$) and $M_{\mathrm{FCC}}(T)$ bifurcate (see figures 3 and 4). It is to be noted that the temperature T_V where $M_{\mathrm{ZFC}}(T)$ and $M_{\mathrm{FCW}}(T)$ merge is actually less than T_{irrv} (see figures 3 and 4) and it has nothing to do with the irreversibility temperature [25]. In the same H–T phase diagram we also mark the low-T high-H irreversible regime with two characteristic temperatures $T_{\mathrm{irrv}1L}$ and $T_{\mathrm{irrv}1U}$. Here $T_{\mathrm{irrv}1L}(H)$ and $T_{\mathrm{irrv}1U}(H)$ represent the lower and upper temperature limit respectively of the observed thermal hysteresis at a particular applied field H (see figure 5).

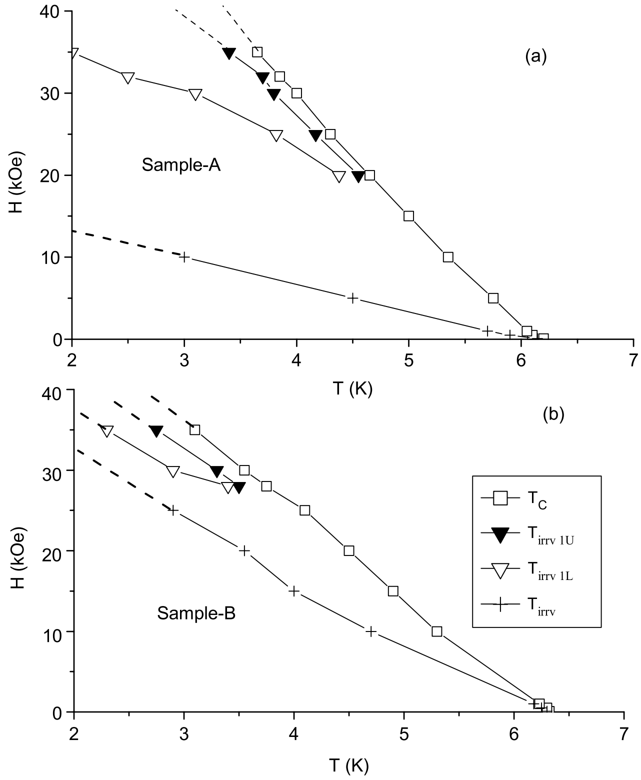

Figure 7. H–T phase diagram for the CeRu$_2$ single crystals (a) sample A, (b) sample B. Here the square symbol represents the superconducting transition temperature (T_C) and '$+$' the irreversibility line $T_{irrv}(H)$. The open (filled) triangle represents the lower (upper) limit T_{irrv1L} (T_{irrv1U}) of the low-T high-H irreversible regime. The dashed lines represent extrapolations from the existing data points.

In sample B the conventional irreversibility regime of the H–T phase diagram bounded by the $T_{irrv}(H)$ line is bigger than that in sample A. On the other hand, the extent of the anomalous low-T high-H irreversible regime, marked by $T_{irrv1L}(H)$ and $T_{irrv1U}(H)$, is distinctly larger in sample A. To highlight this latter observation further, in figure 8 we plot T_C and T_{irrv1L} of sample A and sample B as a function of H. $T_C(H)$ lines for the two samples are extrapolated to obtain the respective $H_{C2}(0)$; these values are in accord with earlier reports [6, 22]. It is to be noted that $H_{C2}(0)$ of the purer sample B is slightly lower than that of the sample A. Such a slightly lower value of H_{C2} and the smaller extent of the low-T high-H irreversible regime in the purer single crystal of CeRu$_2$ have been reported earlier from the isothermal field-dependent measurements [22]. In an earlier study we have investigated the nature of the flux pinning force in a variety of CeRu$_2$ samples including the present single-crystal sample A [20]. A distinct double-peak structure with low pinning force in the intermediate fields was observed for sample A and other samples showing a sharp peak-effect in the isothermal field dependence of magnetization. In contrast, the samples showing weak or no peak-effect had a single peak structure with substantial pinning in the intermediate-field regime [20]. Sample-B clearly belongs to this latter class, and a detailed study of critical current and pinning force of this sample will be part of a future work.

During last ten years a very similar picture of field-induced first-order phase transition in the superconducting mixed state below the $H_{C2}(T)$ line has been proposed in various type-II superconductors both with low T_C [16, 18, 19] and high T_C [17]. Again in all these systems there is no report of temperature-dependent studies confirming the existence of a such a transition, except in a fairly recent study of V$_3$Si [26]. The signature of the phase transition reported in this work on V$_3$Si is much more subtle [26] than that reported

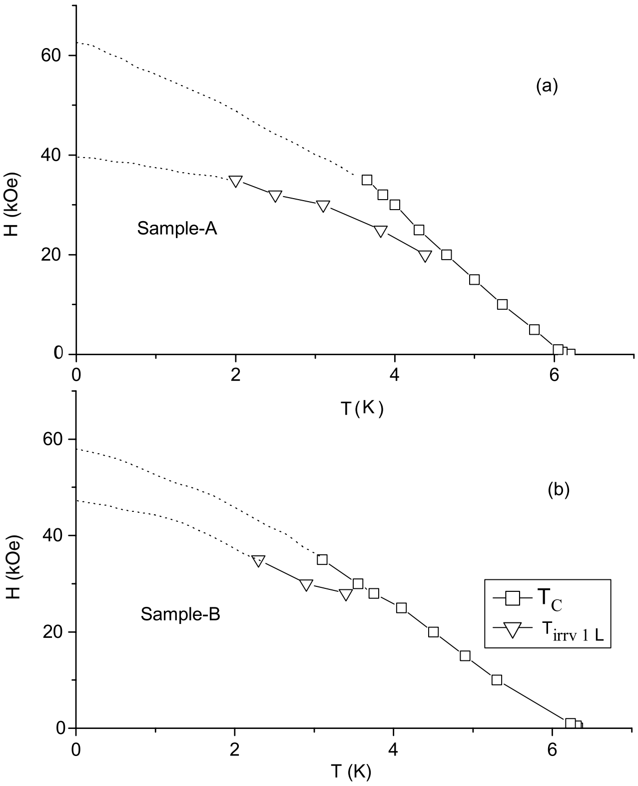

Figure 8. H–T phase diagram for the $CeRu_2$ single crystals (a) sample A, (b) sample B, showing the superconducting transition temperature (T_C) and the lower limit T_{irrv1L} of the low-T high-H irreversible regime. The dashed lines represent extrapolations from the existing data points. The extrapolated values of $H_{C2}(0)$ for the two samples match well with those reported earlier in the literature [6, 22].

here for $CeRu_2$. Moreover, there was no clear correlation between the structure observed in the M–T curve and the peak-effect observed in the isothermal field-dependent study of V_3Si [26]. This is in considerable contrast with the present case of $CeRu_2$, where there is a clear correspondence between the structures observed in the field-dependent and temperature-dependent magnetization studies.

The question now remains: what is the origin of the first-order phase transition in the superconducting mixed state (or vortex state) of $CeRu_2$ (and for that matter in various other type-II superconductors) mentioned above? One possibility is that the competition between thermal energy, pinning energy, and elastic energy of the flux lines can give rise to a crossover from a vortex-matter state with low pinning to a state with high pinning strength [27]. However, there is no proof as yet that such a transformation in pinning strength is indeed a first-order phase transition. On the other hand, there is this possibility of the onset of a Fulde–Ferrel–Larkin–Ovchinnikov (FFLO) state near the $H_{C2}(T)$ line, which is actually through a first-order phase transition [28, 29]. The FFLO phase transition can give rise to a nodal vortex state that is relatively soft, and the existing pinning centres then effectively become strong pinning centres. In the early 1990s experimental observations of the FFLO state were claimed in the uranium-based compound UPd_2Al_3 [30], but this claim was subsequently subjected to some criticisms [31]. It is usually argued that the FFLO state is formed in systems where the orbital motion is strongly suppressed and the upper critical field H_{C2} becomes Pauli limited i.e. $H_{C2orb} > H_P$. Such a favourable situation is expected to be found in quasi-two-dimensional (Q2D) superconductors under a magnetic field parallel to the Q2D plane [32] and in heavy-fermion superconductors with enhanced Pauli paramagnetism and large effective mass [33].

Some experimental supports for the FFLO state have been obtained subsequently in Q2D organic superconductors [34, 35] and in $CeCoIn_5$ heavy-fermion superconductors [36]. And these experimental activities stimulated further theoretical works [37]. The possible existence of an FFLO state in $CeRu_2$, however, still remains a subject of debate [13, 14, 38]. First of all it is not quite clear whether $CeRu_2$ can be considered to be Pauli limited [8, 9] or not [6, 39]. Second, in contradiction to the conventional picture of the FFLO state the suspected signatures of an FFLO state in $CeRu_2$ actually get enhanced with alloying [13, 21, 39]. On a closer inspection, however, it is found that the electronic mean path in the so-called impure alloys of $CeRu_2$ still remains larger than the coherence length [13, 21]. In addition there exists some experimental evidence of a nodal vortex state in $CeRu_2$ [38].

In this context it is worth discussing the rather weak nature of the 'peak-effect' in better-quality single crystals of $CeRu_2$ as observed earlier [22] and in the present work. And, it is an important observation that these purer crystals show higher irreversibility of magnetization in the low-field regime of the superconducting mixed state. Hedo *et al* [22] argued that in $CeRu_2$ there were two kinds of defects: (1) Ru-vacancies, which act like point defects, and (2) planar defects. Of these, the planar defects were not considered to be good pinning centres as the flux lines run away from such defects. According to Hedo *et al* [22] the peak-effect in $CeRu_2$ is mainly due to the Ru-vacancies of which there are definitely more in the relatively impure crystals with low resistivity ratio. However, the idea of relative ineffectiveness of planar defects to pin the flux lines in $CeRu_2$ should be taken with some caution, since the structure of $CeRu_2$ is predominantly a three-dimensional one [22]. If the flux lines run away along one plane, they will be pinned by the planes perpendicular to it. Normal state paramagnetism on the other hand can play some role in the field-temperature response of the superconducting mixed state of $CeRu_2$. Apart from the enhanced Pauli paramagnetism of the normal state of $CeRu_2$, an additional impurity paramagnetic response can arise in $CeRu_2$ due to non-transformed Ce^{3+} ions [5]. Magnetic response can also arise in Ce-based intermetallic compounds from non-magnetic atom disorder [40]. Gschneidner *et al* [40] argued that, although the Ce-atoms in such compounds occupy a periodic lattice, any disorder in the arrangement of the surrounding non-magnetic atoms can cause a variation in the indirect exchange interaction between Ce-ions. Such random magnetic interaction between Ce-ions can give rise to interesting magnetic response including spin-glass-like magnetic ordering at very low temperatures. This mechanism will have strong relevance for $CeRu_2$ especially with the existing suggestion of lattice disorder [22]. Magnetization measurements in the normal state of the present samples A and B reveal a substantial temperature dependence of magnetization, which, however, is not typical Curie–Weiss like. In an earlier study we had actually found that the $CeRu_2$ samples, whose normal state magnetic susceptibilities at 10 K were greater than the corresponding normal state susceptibility $\chi \approx 1.85 \times 10^{-6}$ emu g^{-1} Oe of sample A, showed a clear peak-effect in the isothermal field dependence of magnetization [21]. On the other hand, the samples with lower normal-state magnetic susceptibilities than that of sample A did not show any peak-effect, at least down to 4.5 K [21]. The present higher-purity single-crystal sample B with its normal-state magnetic susceptibility $\chi \approx 1.3 \times 10^{-6}$ emu g^{-1} Oe at 10 K clearly belongs to this latter class. A detailed and comparative analysis of the normal-state magnetic properties of samples A and B with the suppression of the superconducting state by applying high magnetic fields (up to 80 kOe) will be published in the future.

It was argued by Kadowaki *et al* [8] that the paramagnetism in $CeRu_2$ can reduce the condensation energy by an amount $\pi\xi^2\chi_{\mathrm{spin}}h^2/2$, where χ_{spin} is the paramagnetic susceptibility, ξ is the coherence length, and h is the magnetic field inside the vortex core. This latter energy, which can be about 1/3 of the condensation energy, can be a cause of the large reversible (H, T) regime in $CeRu_2$. The role of paramagnetism in $CeRu_2$ is quite visible around $H_{C2}(T)$

in both samples A and B studied here. There is a clear presence of a paramagnetic mixed state in both the samples in the low-T isothermal M–H curves (see figures 2 and 3) and high-H constant-field M–T curves (figures 5 and 6); the magnetization is clearly positive near H_{C2} and T_C. Also, the hysteresis bubble in the M–T measurements (indicative of the phase transition) is entirely confined to the paramagnetic regime in both samples. In earlier studies a distinct paramagnetic mixed state was also observed near H_{C2} even in a single crystal with resistivity ratio of 270 [22]. It is worth recalling here that in the original work of Fulde and Ferrel [28] paramagnetic impurities in a type-II superconductor were thought to be instrumental for the onset of the FFLO state with nodal vortices. While this information is pertinent to the superconducting mixed state of $CeRu_2$, it should not bias a reader towards the existence of an FFLO state in $CeRu_2$. The concrete proof in this regard (or against it) is yet to be established.

In conclusion, there are definite evidences of a temperature and magnetic field-induced vortex-matter phase transition in the superconducting mixed state of $CeRu_2$. The indication of this transition originally came through the isothermal field variation studies of magnetization, and it took quite a bit of time to establish the first-order nature of this vortex-matter phase transition [13, 14]. There was no report so far on the existence of a temperature-induced first-order phase transition in the superconducting mixed state of $CeRu_2$. The present study now shows that this vortex-matter phase transition can also be induced by temperature, and establishes further the first-order nature of this transition. This study also highlights the rather intriguing behaviour of the superconducting mixed state of $CeRu_2$ as a function of defects. Very pure single crystals have larger low-field pinning force in the superconducting mixed state; on the other hand, the phase transition in the high-H regime and the associated peak effect seem to be less robust in such crystals. These latter effects are very prominent in the relatively impure single crystals and exist even in a higher-T regime (than in the purer single crystal). But the pinning effect in the low-field regime is perceptibly weaker in these low-purity crystals. These observations are definitely counterintuitive and clearly point out that competition between thermal energy, pinning energy and elastic energy alone cannot probably explain the low-temperature high-field first-order vortex-matter transition in the superconducting mixed state of $CeRu_2$. Paramagnetic properties of the normal state of $CeRu_2$ (be it extrinsic or intrinsic) seem to play an important role in the observed response of the superconducting mixed state.

Acknowledgments

We thank Professors A D Huxley and L E DeLong for providing us with the single crystals of $CeRu_2$ used in this study.

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