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# Evidence for supercooling across a vortex-matter phase transition: studies on $CeRu_2$ and $Bi_2Sr_2CaCu_2O_8$

**P CHADDAH* and S B ROY**

Low Temperature Physics Group, Centre for Advanced Technology, Indore 452 013, India

**Abstract. We present DC magnetization data indicating a first-order phase transition in the vortex state of $CeRu_2$, with the higher entropy phase exhibiting enhanced pinning. Minor hysteresis loops show evidence of supercooling of the higher entropy phase as the phase boundary is crossed both isothermally as well as at constant field. These features are shown to be absent across the Bragg-glass to vortex-glass transition in $Bi_2Sr_2CaCu_2O_8$. The supercooling is more persistent in the constant field case.**

**Keywords. Vortex-matter transition; supercooling; $CeRu_2$; $Bi_2Sr_2CaCu_2O_8$.**

## 1. Introduction

The most extensively studied phase transition in vortex matter is the first-order melting of the vortex lattice in high-$T_C$ superconductors (HTSC) (see Blatter 1997). In this paper we concentrate on possible phase transitions below this melting line. In recent years the phase diagram of vortices in HTSC with weak disorder has been studied in great detail theoretically. The low-field Bragg-glass phase is expected to undergo a second-order or continuous phase transition, with increasing magnetic field, to a vortex-glass phase—with the driving mechanism being the proliferation of dislocation loops (Ertas and Nelson 1996). Recent experimental studies on various HTSC (Khaykovich *et al* 1996; Deligiannis *et al* 1997; Giller *et al* 1997) correlate onset of the peak effect with this continuous phase transition.

A first-order phase transition in the vortex lattice of paramagnetic superconductors, at high fields, was proposed over three decades ago (Fulde and Ferrel 1964; Larkin and Ovchinnikov 1965; Gruenberg and Gunther 1966). The anomalous peak effect observed in $UPd_2Al_3$ and $CeRu_2$ has, in various works, been discussed in this context (see for e.g. Modler *et al* 1996; Roy and Chaddah 1997a). In a generalized version of the original theory (Tachiki *et al* 1996; Takahashi *et al* 1996), the vortices are segmented into short strings with a concomitant enhancement of pinning. The onset of peak effect in these materials has been accordingly investigated as driven by a first-order phase transition to this generalized Fulde–Ferrel–Larkin–Ovchinnikov (GFFLO) state.

In this paper we shall present our detailed studies on the paramagnetic superconductor $CeRu_2$, where the anomalous magnetization just below $H_{C2}$ was discovered by one of us (Roy 1992). We have studied a large number of doped and undoped samples, and the results to be discussed here are generic to all samples with a normal state susceptibility above a threshold value (Roy and Chaddah 1997a, 1998). We shall also present our similar comparative studies on $Bi_2Sr_2CaCu_2O_8$ (hereafter referred to as Bi-2212), where the Bragg-glass to vortex-glass transition concomitant with the onset of peak effect is second-order or at most weakly first-order (Khaykovich *et al* 1996).

## 2. Isothermal variation of magnetic field

Before proceeding with the anomalous peak effect, we wish to record two striking features of $CeRu_2$ that have not yet been addressed in detail in literature. First, addition of magnetic dopants at Ce site raises $T_C$ as well as $H_{C2}$ (Wilhelm and Hillenbrand 1971; Roy and Coles 1990). Second, the *M–H* curve just below $H_{C2}$ is reversible but approaches the normal state magnetization from above (Yagasaki *et al* 1993; Roy and Coles 1994; Nakama *et al* 1995). Both these features are anomalous and understanding them may tell us more about the microscopics of superconductivity in $CeRu_2$.

Figure 1 shows a typical *M–H* curve of $CeRu_2$ (this data is on a 5% Nd-doped sample at 4·5 K). There are two distinct regions of irreversible magnetization; a low-field region I (below 7 kOe) and a high-field region II (around 30 kOe). Roy *et al* (1998d) have argued that while the irreversibility and concomitant metastability in region I, like that of other hard type-II superconductors (including Bi-2212), is akin to that of spin-glasses, the metastability in region II is akin to that of random-field Ising systems. The main characteristics underlying this differentiation are (i) $M_{FC} > M_{eq}$ in region I, while $M_{FC} < M_{eq}$ in region II; and (ii) $dM_{FC}/dt$ is negative in region I but positive in region II.

*Author for correspondence

Minor hysteresis loops (MHLs), measured at closely spaced field intervals, provide a method of understanding the nature of magnetic irreversibility in superconductors. In particular, one can distinguish between surface effects and bulk irreversibility. Further, one can ascertain whether the irreversibility is consistent with Bean's critical state model (CSM) because the behaviour of MHLs therein is well documented (Chaddah *et al* 1992).

The MHLs in Bi-2212 (Roy *et al* 1998b) show a continuously varying initial slope, indicating the absence of surface-driven hysteresis. They also merge with the envelope hysteresis curve as predicted by the CSM. Pradhan *et al* (1998) have similarly reported MHLs in the peak effect region of $NdBa_2Cu_3O_7$ consistent with the CSM. Even in $CeRu_2$, MHLs are consistent with the CSM in region I. In region II also, the MHLs are non-linear throughout, ruling out surface effects. The MHLs initiated from the lower envelope curve just above the onset of region II, however, do not show the expected merger with the upper envelope curve. As shown by Roy and Chaddah (1997b) the saturated value of the MHL at say 26·5 kOe further depends on whether the MHL was initiated from 27 kOe or from 29 kOe (see figure 2). Roy and Chaddah (1997b) and Roy *et al* (1998a) have argued that this indicates the formation of a new vortex phase (say phase X) at the onset of region II. This phase X, in contrast to the vortex-glass phase formed at the onset of the peak effect in Bi-2212, supercools on reducing the field.

We show in figure 3, a schematic phase diagram of superconducting $CeRu_2$ (Roy and Chaddah 1996; Dilley and Maple 1997) in which $H_a^*$ gives the onset of the peak effect or region II, and $H_{pk}$ is the field value in region II where the magnetization hysteresis shows a peak. What the data of figure 2 clarifies is that MHLs show multivaluedness of saturation magnetization $M_S$ as the field is raised above $H_a^*$ and before it reaches $H_{pk}$. The transition to phase X is thus initiated at $H_a^*$. Roy and Chaddah (1997c) and Roy *et al* (1998a) have argued that sample geometry may cause the transition to have a width of a few kOe. The multivaluedness of $M_S$ results from supercooling of states in which different extents of phase X have formed. Such a supercooling is not seen from a study of MHLs across the continuous phase transition, from vortex-glass to Bragg-glass phase, in Bi-2212 (Roy *et al* 1998b) and in $NdBa_2Cu_3O_7$ (Pradhan *et al* 1998).

Measurement of MHLs at closely spaced field intervals also allows one to estimate artifacts that may be introduced in data taken on a SQUID-magnetometer because of the sample being moved through the inhomogeneous field of the magnet (Roy and Chaddah 1996). We have estimated that these artifacts are insignificant in our data (Roy *et al* 1998b). This artifact is absent in data taken on a vibrating-sample-magnetometer, and we have confirmed the anomalous nature of MHLs in region II, in both pure and doped $CeRu_2$, through collaborative measurements using a VSM (Chaudhary *et al* 1998b).

## 3. Variation of temperature

We now discuss signatures of supercooling on crossing from region II to region I in constant field.

Various thermomagnetic histories had been followed by Steingart *et al* (1973), while studying transport $J_C$ in a strained Nb crystal exhibiting peak effect. They found that $J_C$ measured in the peak effect region was highest on cooling in constant field, next when field

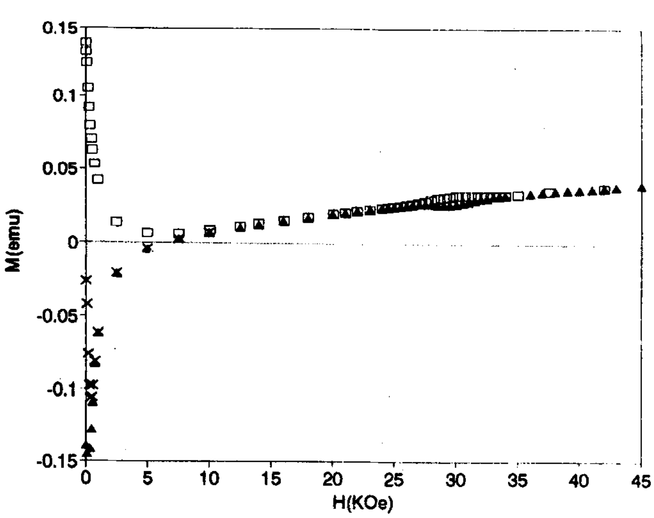


**Figure 1.** $M$ vs $H$ plot of $(Ce_{0.95}Nd_{0.05})$ $Ru_2$ at $T = 4{\cdot}5$ K. Cross is virgin, square is descending and triangle is ascending magnetization cycles.

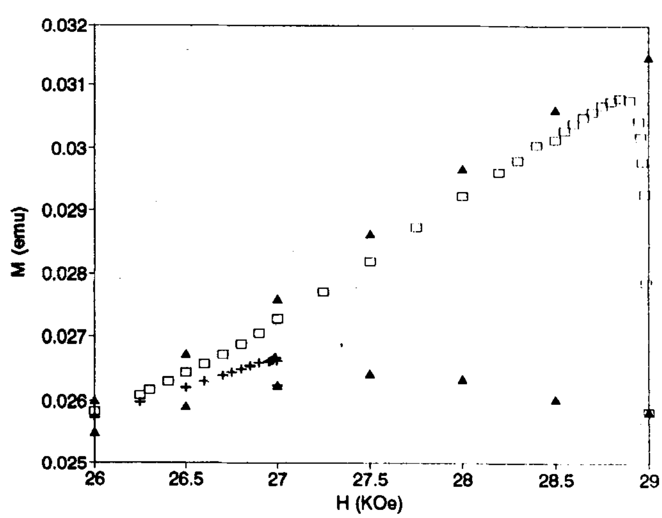


**Figure 2.** MHL initiated from the lower envelope curve at 27 kOe (cross) and 29 kOe (square) do not merge with the envelope curve (triangles) and show different saturated values at 26·5 kOe. Sample and temperature is same as in figure 1.

was lowered isothermally from above $H_{C2}$, and lowest when field was raised isothermally from zero. While they did not consider a phase transition in the vortex state, we view the qualitative similarity between their isothermal transport data and our isothermal magnetization data as significant. While explaining the different $J_C$s obtained under different histories as due to synchronized pinning, Steingart *et al* (1973) had argued that the readjustment of vortex spacing is least in field-cooled case. The energy change experienced by vortices is thus least in this case. Supercooling of phase X can also be expected to persist farthest when the energy change of vortices is least. The arguments of Steingart *et al* (1973) thus prompted us to access region I from region II by cooling in constant field. As explained earlier, multi-valuedness of saturation magnetization of MHLs is attributed to supercooling of phase X. Roy *et al* (1998c) found no supercooling in Bi-2212 on field-cooling, but supercooling was seen in $CeRu_2$ and persisted to much lower fields. Specifically, supercooling was not seen below 20 kOe at 5 K on isothermal field reduction in Nd-doped $CeRu_2$ (Roy and Chaddah 1997b), but persisted down to 13 kOe and 5 K on field-cooling (Roy *et al* 1998c).

Finally, field-cooling failed to show multivaluedness or supercooling above a critical temperature $T^*$, indicating a critical point in the schematic phase diagram.

## 4. Failure-test for a first-order transition

We now discuss a true thermodynamic signature of the isothermal transition at $H_a^*$, being a first-order transition. There has recently been quite some discussion on the observation of a first-order transition of the vortex lattice (Welp *et al* 1996). The equilibrium magnetization $M_{eq}$ is a thermodynamic quantity whereas resistivity is not. In clean single crystals of HTSC, the melting transition shows a clear change in $M_{eq}$ vs $H$ since the $M$–$H$ curve is reversible. In $CeRu_2$, we have to infer $M_{eq}$ from a hysteretic $M$–$H$ (Chaddah *et al* 1998), which is further complicated by supercooling. We have accordingly used the MHLs to estimate $M_{eq}$ (Roy *et al* 1998a, b).

We first note that the field $H_a^*$ at which the transition is seen rises as $T$ falls. Further the high field phase is also the high temperature phase and therefore has higher entropy. Taken together with the Clausius–Clapeyron relation, the volume of the vortex state must fall or $M_{eq}$ must rise as the field is raised isothermally across $H_a^*$. A drop in $M_{eq}$ at $H_a^*$ would imply a failure of the first-order transition hypothesis. We have measured $M_{eq}$ vs $H$ as an essential failure test.

We show in figure 4 the results of our measurement on a pure sample of $CeRu_2$. A perceptible rise at $H_a^*$ is also seen in doped $CeRu_2$ (Roy and Chaddah 1997c). We have made similar measurements at other temperatures (Roy *et al* 1998a) and in figure 5 we plot the field-dependence of the rise in the $M_{eq}$ over the extrapolated value. We have found that the jump in $M_{eq}$ drops to zero as $T$ rises from $0{\cdot}75\ T_C$ to $0{\cdot}9\ T_C$, consistent with the existence of a critical point at $T^*$. The jumps shown in figure 5 have a width $\Delta H/H_a^*$ around 0·1, whereas one expects a sudden rise in a first-order transition. In the evidence for flux lattice melting in $YBa_2Cu_3O_7$, presented by Welp *et al* (1996) in their figures 1 and 2, the rise in $M_{eq}$ also occurs over about 2 kOe even though their $M$–$H$ data is reversible. The results of Zeldov *et al*

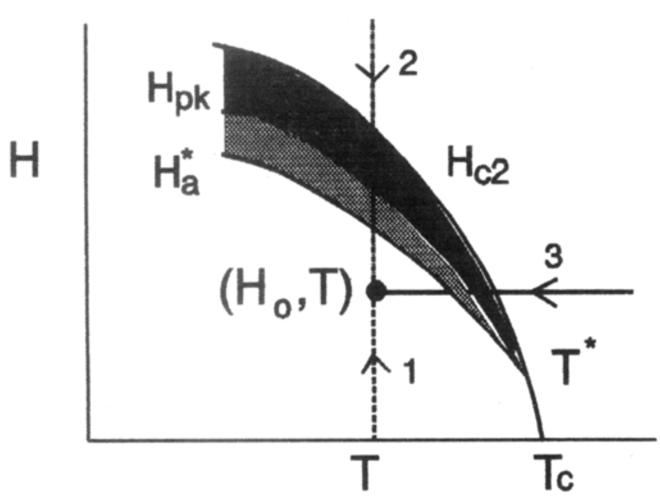


**Figure 3.** Schematic phase diagram of superconducting $CeRu_2$. Paths indicated by 2 and 3 correspond to isothermal field reduction and to field-cooling through the high-entropy phase. A critical point is indicated at $T^*$, above which this phase does not exist.

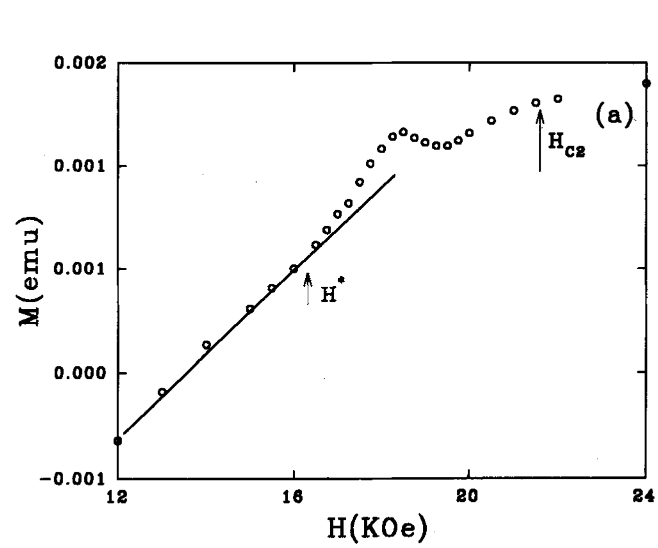


**Figure 4.** Equilibrium magnetization ($M_{eq}$) vs field ($H$) plot for a pure $CeRu_2$ sample at 4·5 K. Note the distinct rise in $M_{eq}$ at $H_a^* = 16{\cdot}5$ kOe.

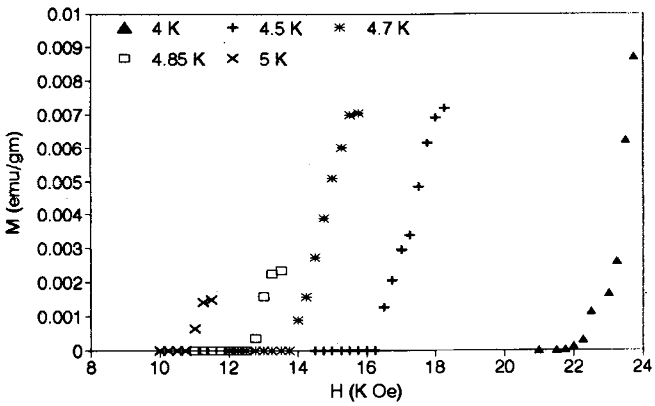


**Figure 5.** Field dependence of the jump in $M_{eq}$ at various temperatures.

(1995) show that such a width in a first-order transition can be understood as a sample shape artifact in global magnetization measurements.

## 5. Conclusion

We have presented DC magnetization data that indicates a phase transition in the vortex state of $CeRu_2$, with the higher entropy phase exhibiting enhanced pinning. We have shown evidence of supercooling across the phase boundary as field is lowered isothermally. We have then shown evidence of more persistent supercooling when the sample is cooled across the phase boundary in constant field. This more persistent supercooling is consistent with the smaller vortex motion anticipated in the field-cooled case (Steingart *et al* 1973). We have shown that in both the histories in which supercooling is seen in $CeRu_2$, no supercooling is seen across the vortex-glass to Bragg-glass transition in Bi-2212. We have shown that the $M_{eq}$ vs $H$ for $CeRu_2$ is not inconsistent with a first-order transition of the vortex lattice.

We would finally wish to speculate on the origin of this first-order transition. The Bragg-glass to vortex-glass transition is believed to be driven by the gradual proliferation of dislocation loops favoured by point disorder (Ertas and Nelson 1996). The segmentation of vortices in the GFFLO state could also cause a proliferation of dislocation loops. Since the formation of GFFLO state, with the concomitant segmentation of vortices, is through a first-order transition, the proliferation of dislocation loops would be sudden and not gradual as in HTSC. Our recent correlation (Chaudhary *et al* 1998a) of irreversibility magnitudes in regions I and II amongst various samples of $CeRu_2$, also indicates the possible role of magnetic impurities.

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