Manifestation of history dependent critical currents via dc and ac magnetisation measurements in single crystals of $CeRu_2$ and $2H - NbSe_2$

G. Ravikumar, V.C. Sahni, P.K. Mishra and T.V. Chandrasekhar Rao

Technical Physics and Prototype Engineering Division, Bhabha Atomic Research Centre, Mumbai 400 085, INDIA

S.S. Banerjee, A.K. Grover, S. Ramakrishnan and S. Bhattacharya

Department of Condensed Matter Physics and Material Science, Tata Institute of Fundamental Research, Mumbai -400 005, INDIA

M.J. Higgins

NEC Research Institute,4 Independence way, Princeton, NJ 08540

E. Yamamoto [1], Y. Haga[1], M. Hedo [2], Y. Inada[2] and Y.Onuki[1,2]

[1]Faculty of Science, Osaka University, Toyonaka 560, JAPAN

[2]Advanced Science Research centre, Japan Atomic Energy Research Institute, Tokai, Ibaraki 319-11, JAPAN

A study of path dependent effects in single crystals of $CeRu_2$ and $2H - NbSe_2$ show that critical current density J_c of the vortex state depends on its thermomagnetic history over a very large part of (H, T) parameter space. The path dependence in J_c is absent above the peak position (i.e., $H > H_p$) of the peak effect region, which we believe identifies the complete loss of order in the vortex structure. The highly disordered FC state can be healed into a relatively ordered vortex lattice by subjecting it to a large enough change in dc field (few tens of Oe) or by shaking the FC state with sufficient ac field (few Oe).

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Investigating structure of vortex lattice or flux line lattice (FLL) in the mixed state of type II superconductors continues to be of intense interest. Recent theoretical studies have postulated various glassy states in FLL arising from quenched disorder and thermal fluctuations [1]. Experimental efforts have been focused on detection and characterisation of such states. The appearance of the Peak Effect (PE) in some systems involving an anomalous enhancement of critical current density J_c in close proximity of the softening/melting in their FLL [2–4] - has been explained in terms of a loss of spatial order in FLL [5]. But the precise nature of this loss of order and its relationship to the glassy state of FLL are topics of current debate.

In disordered magnetic systems such as spin glasses [6], one encounters the appearance of thermomagnetic history effects. The temperature below which magnetisation values under zero field cooled (ZFC) and field cooled (FC) conditions differ is usually identified as the spin glass transition temperature T_g [6]. We find analogous manifestations in the magnetic behaviour in weakly pinned superconducting systems, viz., $2H - NbSe_2$(T_c=6.1K) [7] and $CeRu_2$(T_c=6.3K) [8], as reflected in their J_c values. This difference in the FC and ZFC response, we believe, reflects the different extents of FLL correlations in these states, and is seen to persist upto the peak position of PE. In this sense the locus of PE in H-T plane may be regarded as the counterpart of spin glass transition temperature. The results of dc and ac magnetisation

experiments in the two superconductors are presented so as to elucidate history effects well before, just prior to, during and after the occurence of peak of the PE phenomenon. Our results *inter alia* add a newer facet to the well known Critical State Model (CSM) [9] which postulates a unique critical current density J_c for the vortex state for a given field (H) and temperature (T).

We recall that hysteresis in magnetisation is related to $J_c(H)$ [10]. In isothermal magnetisation measurements, this single valued J_c translates into a generic magnetisation hystersis loop (see Fig.1) such that the *forward* and *reverse* branches of the magnetisation curve define an envelope [10], within which lie all the magnetisation values that can be measured at the given temperature along various paths with different thermomagnetic histories [11]. For instance, Fig.1 schematically illustrates that the FC magnetisation curve generated by decreasing the field after cooling the sample in a dc field eventually merges into the reverse magnetisation branch. *The new result of our experiments is that the magnetisation curve, originating from a given FC state, need not always be confined within the generic hysteresis loop.* We argue that the observed new behaviour elucidates the existence of multivalued nature of $J_c(H, T)$, i.e., the critical current density of vortex state at a given (H, T) depends on its thermomagnetic history. Our inference by an equilibrium dc magnetisation technique strengthens an earlier conclusion from non-equilibrium transport studies on $2H - NbSe_2$ by Henderson et al [7], who measured that

the transport J_c in ZFC state is considerably lower than that in the FC state for fields below the peak field H_p.

AC susceptibility measurements in superimposed dc fields were performed on a home built ac susceptometer [12]. The dc magnetisation measurements have been performed using Quantum Design (QD) Inc. (Model MPMS) SQUID magnetometer. The single crystals of cubic $CeRu_2$ and hexagonal $2H-NbSe_2$ were mounted on the sample holder such that the field is parallel to cube edge and c-axis, respectively. Usually, the measurement of magnetic moment m in the MPMS SQUID magnetometer involves sample motion along the pickup coil array in the second derivative configuration, over a scan length $2l$. The magnetic moment m is obtained by fitting the *sample response* measured over $-l < z < l$ to the form,

$$V = a + bz + mc\phi(z - z_0), \qquad (1)$$

where,

$$\phi(z) = (\mu_0 R^2/2) \left[-[R^2 + (z+Z)^2]^{-3/2} \right.$$

$$\left. +2[R^2 + z^2]^{-3/2} - [R^2 + (z-Z)^2]^{-3/2} \right] \qquad (2)$$

Here, a, b and z_0 account for constant offset, linear drift and possible off-centering of the sample respectively. R (= 0.97 cm) is the radius and $2Z$ (= 3.038 cm) is the distance between the two outer turns of the pick-up coil array. z is the sample distance from the centre of the pickup coil array and c is the calibration factor. This analysis implicitly assumes that m is constant along the scan length and therefore independent of z. But, as described in Ref. [13], when a superconducting sample, which exhibits PE, is moved in an inhomogeneous external field, its magnetic moment can become strongly position dependent, leading to spurious experimental artefacts in the data. Thus, an appropriate method needs to be devised to obtain magnetisation values which are free from such artefacts. We have done this, by analysing the raw data using a new procedure that can be termed as *half-scan* technique and its salient features are detailed below.

In 5.5 Tesla QD MPMS model, on either side of the centre of the magnet [14] the field due to the superconducting solenoid monotonically decreases along the axial direction. The central idea of the *half scan* technique is to record the *sample response* by moving it over that part of the axis so that the sample does not experience field excursions. On the *forward* magnetisation curve, this is accomplished by recording the sample response *only* between $z = -l$ and $z = 0$. As the magnetisation of the sample stays nearly constant for $-l < z < 0$, we can fit this data to Eqn. 1 and obtain magnetic moment m on the *forward* magnetisation curve. As illustrated in Fig.2a, the SQUID response in the conventional measurement (spanning $-l$ to l) fits very poorly to the ideal dipolar

response given by Eqn.1. On the other hand the *half-scan* response measured between $z = -2$ cm and $z = 0$ gives an excellent fit to Eqn.1. The SQUID response of $2H-NbSe_2$ single crystal, shown in Fig.2 is measured at 4.5K in a field of 8 kOe, i.e., very close to peak field H_p. Similarly, to obtain the magnetic moment on the *reverse* magnetisation curve, the sample is initially positioned at $z = 0$ (i.e., where the field is maximum along the axis of the solenoid). The SQUID response shown in Fig.2b is recorded by moving the sample between $z = 0$ and $z = l$. Magnetic moment is then obtained by fitting this response to Eqn.1. The SQUID responses shown in Fig.2(a) and (b) have been compensated for the offset a and the drift bz (cf. Eqn. 1).

Figs. 3 and 4 summarize the central results of magnetisation hysteresis and ac susceptibility experiments in crystals of $2H-NbSe_2$ ($2 \times 2 \times 0.4$ mm^3) and $CeRu_2$ ($3 \times 1.5 \times 0.8$ mm^3). (It may be stated here that the $2H-NbSe_2$ crystal is from the same batch as was used by Henderson et al [7] and $CeRu_2$ crystal is the one used for de-Haas van Alphen studies earlier [15]. As mentioned earlier, both these superconducting systems are weakly pinned and the crystal pieces chosen for present measurements have comparable levels of quenched disorder in them [16]). Figs. 3a and 4a display the magnetisation hysteresis loops in the PE regime of $2H-NbSe_2$ and $CeRu_2$ respectively. The pronounced increase in the hysteresis in the PE region of both $2H-NbSe_2$ and $CeRu_2$ signify the anomalous increase in the critical current density at the onset of PE. Figs. 3a and 4a, also, show the magnetisation curves measured in reducing fields, after having cooled the samples in the pre-selected magnetic fields to a given temperature. The pre-selected field cooled magnetisation states can be identified by filled diamonds lying on the dashed line in Figs. 3a and 4a. Magnetisation of the FC sample in reducing magnetic field is measured in the same way as the *reverse* magnetisation curve is generated. In ac susceptibility measurements, the PE manifests via an enhanced (shielding) diamagnetic response. Fig.3(b) shows the plot of in-phase ac susceptibility (χ') vs H in ZFC and FC states in $2H-NbSe_2$ crystal at 5.1 K and Fig. 4(b) shows similar results for $CeRu_2$ crystal at 4.5 K. The χ' data points in FC states were measured after cooling down the sample in a given H to the respective temperatures from the normal state.

It can be seen in Figs.3(a) and 4(a) that the magnetisation curve measured on field cooling in $H > H_p$ readily merges with the usual *reverse* magnetisation curve. This can be well understood within the framework of conventional Critical State Model [9,10], which assumes that J_c is single valued function of (H,T) (see Fig. 1). However, when field cooled in $H < H_p$, the magnetisation values obtained by reducing the external field initially **overshoot** the *reverse* magnetisation curve (see Fig. 3a and Fig. 4a). On further reducing the field, the magnetisation values fall sharply and FC magnetisation curve merges into the usual *reverse* magnetisation hysteresis branch. The first observation that the magnetisation val-

ues initially go beyond the conventional hysteresis loop is a clear indication of J_c at a given H in the FC state (J_c^{FC}) being larger than that for the vortex state at the same H value on the usual *reverse* magnetisation branch. The later observation that the FC magnetisation curve eventually merges into the *reverse* magnetisation branch implies that the FC vortex state transforms to a more ordered ZFC like state as the vortex state adjusts to a large enough change (10^2 Oe for $CeRu_2$ and 10 Oe for 2H-NbSe$_2$) in the external *dc* field. A neutron study [17] on a crystal of $CeRu_2$ had shown that the FC state far below the PE region comprised much more finely divided blocks than that in the ZFC state. Keeping this in view, on the basis of present results, it may be stated that the finely divided FC vortex state heals to the more ordered ZFC state in response to changes induced by large external field variation.

The *ac* susceptibility data in Figs. 3(b) and 4(b) corroborate the above stated conclusions. As per a CSM result [9], $\chi' = -1 + \alpha h_{ac}/J_c$, where α is a shape and size dependent parameter and h_{ac} is the *ac* field amplitude, the higher diamagnetic response in the FC state as compared to that for the ZFC state, reflects larger J_c in the former state. The history dependence in χ' response ceases above the peak position of the PE region. Also, at very low fields ($H < 1kOe$), the difference between FC and ZFC χ' response is seen to decrease, consistent with transport J_c measurements of Henderson et al [7].

In the Larkin-Ovchinikov [18] description of pinning in superconductors, $J_c \propto V_c^{-1/2}$, where V_c is the volume of a Larkin domain within which flux lines remain correlated. Smaller J_c in the ZFC state [7,19], therefore, corresponds to a more ordered FLL than in the case of FC state. For $H > H_p$, flux lines form a quasi-pinned state, which appears to be independent of how the state is approached in (H,T) space. For $H < H_p$, the larger J_c^{FC} can therefore be attributed to the formation of a more finely divided disordered state, with concomitant more pinning. While subjecting the FC state to a decrease in the external field, this state eventually goes over into a relatively more ordered ZFC state with a larger V_c, as manifested by a steep fall in the magnetisation values (after overshooting the *reverse* magnetisation branch). A change from a disordered state to more ordered vortex state can also be brought about by other kinds of perturbations as well. For example, in our *ac* measurements, we observed that the large (shielding) diamagnetism of FC state suddenly collapses to that of the ZFC state on increasing the *ac* field amplitude momentarily to about 5 Oe. Although by no means obvious this way of *annealing* away the disorder of the FLL in the FC case is akin to annealing by a passage of large transport current [7].

$CeRu_2$ and $2H - NbSe_2$ are very dissimilar superconducting systems as regards their microscopic physics; the former is a mixed valent system whereas the latter is a layered chalcogenide which exhibits charge density wave behaviour in its normal state. In the context of vortex state of superconductors, the PE phenomena in weakly pinned samples [2,7,8] of these two systems has been in current focus (apparently) due to different reasons. PE in $CeRu_2$ has (often) been considered to relate to realization of Generalized Fulde-Ferrel Larkin Ovchinikov (GFFLO) state [8], whereas in very clean samples of $2H - NbSe_2$, PE is ascribed to FLL softening [2]. Since normal state paramagnetism of $2H - NbSe_2$ is small, it could not be a serious candidate for the occurence of GF-FLO state. The present findings, that the experimental features of the mixed state prior to and across the PE region of these two systems follow identical course, would support the view that their behaviour in the PE regime presumably reflects same generic physical phenomenon that occurs in the mixed state in a weakly pinned flux line lattice while approaching H_{c2}.

To conclude, we have demonstrated through *dc* and *ac* magnetisation measurements with a new *half-scan* technique, in the mixed state of $CeRu_2$ and $2H - NbSe_2$, that there are sizable thermomagnetic history effects in their critical currents below H_p, where the peak of the PE occurs. We have shown that these effects imply a more finely divided disordered vortex arrangement for the FC state, as compared to that for the ZFC state. It should be noted that the critical current, remains finite above H_p. This suggests that the glassy state above H_p is weakly pinned and a change to completely unpinned state does not appear until the higher field H_{irr}. The implication of these results with respect to the occurence of PE behaviour, fishtail (second peak), etc., in the Cuprate superconductors, such as, YBCO and BSCCO, remains an interesting topic for further investigations.

*Present and permanent address: NEC Research Institute, 4 Independence Way, Princeton, NJ 08540, USA.

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FIG. 1. A schematic showing magnetisation hysteresis curves in an irreversible type II superconductor. *Forward* and *reverse* magnetisation branches corresponding to increasing and decreasing field cycles. A magnetisation curve measured during reducing field cycle after cooling in a field is indicated as field cooled (FC) curve. M_{FC} denotes the FC magnetisation value.

FIG. 2. (a)(○) SQUID response of the sample in the PE region, on the *forward* magnetisation curve using a conventional full symmetric scan of 4cm length. The corresponding fit to Eqn.1 is shown by a dotted line. The *half-scan* response for $-2cm < z < 0$ (●) and its fit to Eqn.1 (continuous line) is also shown. (b) The SQUID response from 0 to 2 cm for the *reverse* case is shown along with the corresponding fit to Eqn.1 after compensating for the offset a and linear drift bz.

FIG. 3. (a) A portion of the magnetisation hysteresis curve (encompassing the PE region) recorded at 5.1K for $H \parallel c$ using the *half-scan* technique in a $2H - NbSe_2$ crystal. Also shown are the magnetisation values recorded while decreasing the field after cooling the sample in (pre-selected) different external fields. The initial M_{FC} values are identified by filled diamonds lying on the dashed curve. Each FC magnetisation curve initiates from a different M_{FC} value. (b) AC susceptibility measured with $h_{ac} = 0.5$ Oe at $f = 211$ Hz, for $H \parallel c$ at 5.1K (i) after cooling the sample in zero field (ZFC) and (ii) after cooling the sample different fields each time (FC). $\chi'(H)$ values are normalized to $\chi'(0)$

FIG. 4. (a) A portion of the magnetisation hysteresis curve of $CeRu_2$ recorded at 4.5K for $H \parallel [100]$ using the *half-scan* technique. The FC magnetisation curves originating from different M_{FC} values are also shown. (b) AC susceptibility for $H \parallel [100]$ in both ZFC and FC modes as described in the caption of Fig.3b.
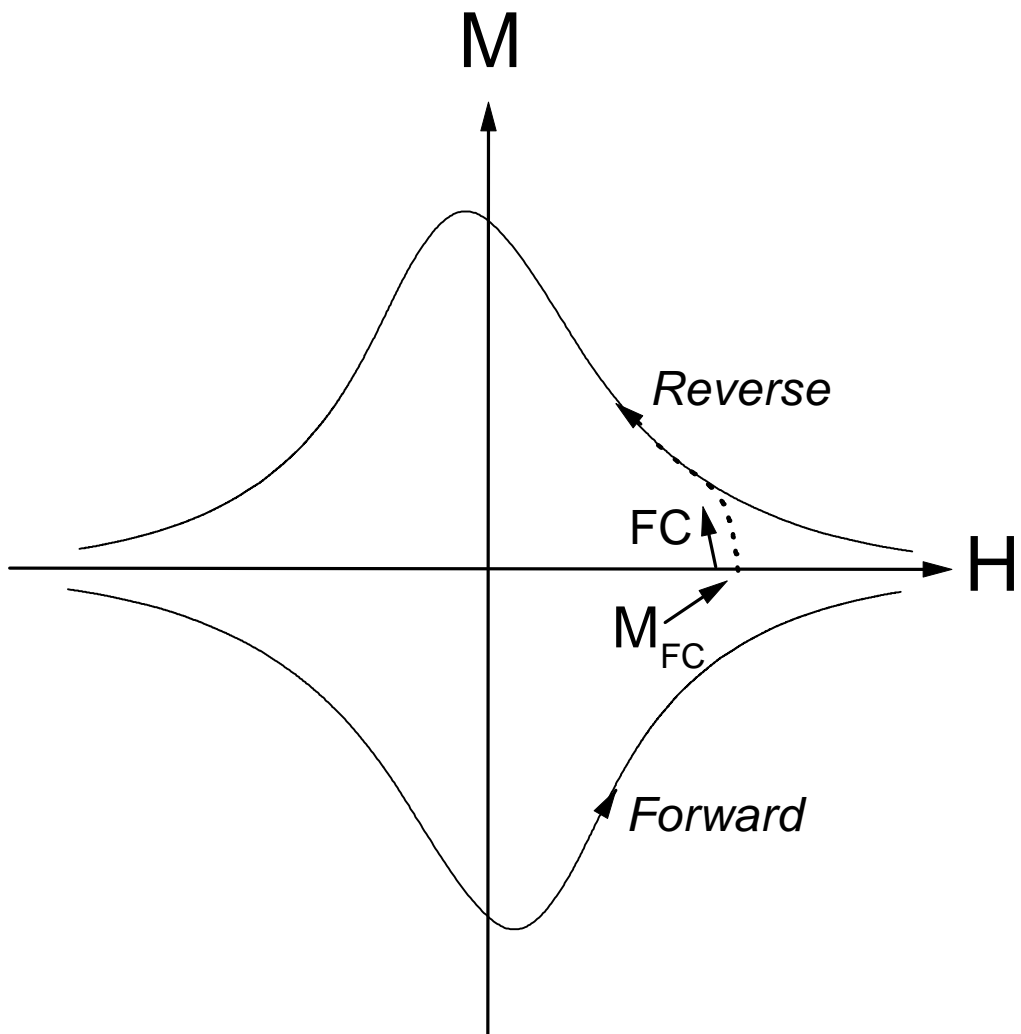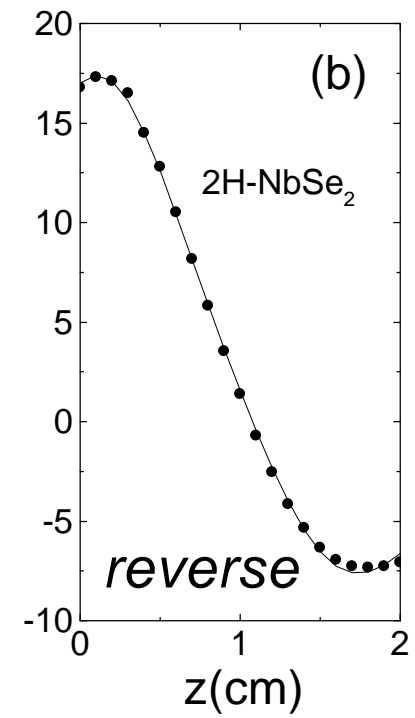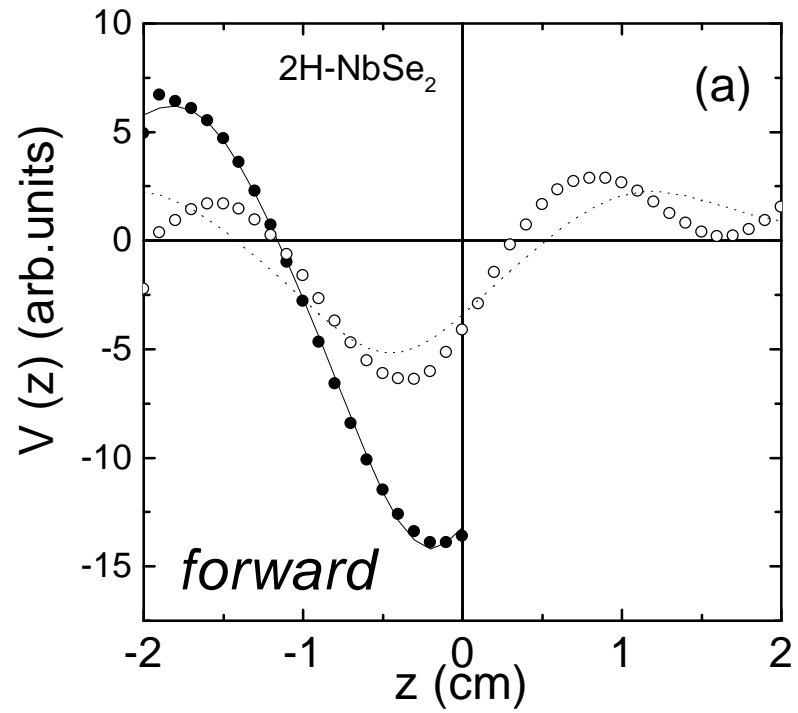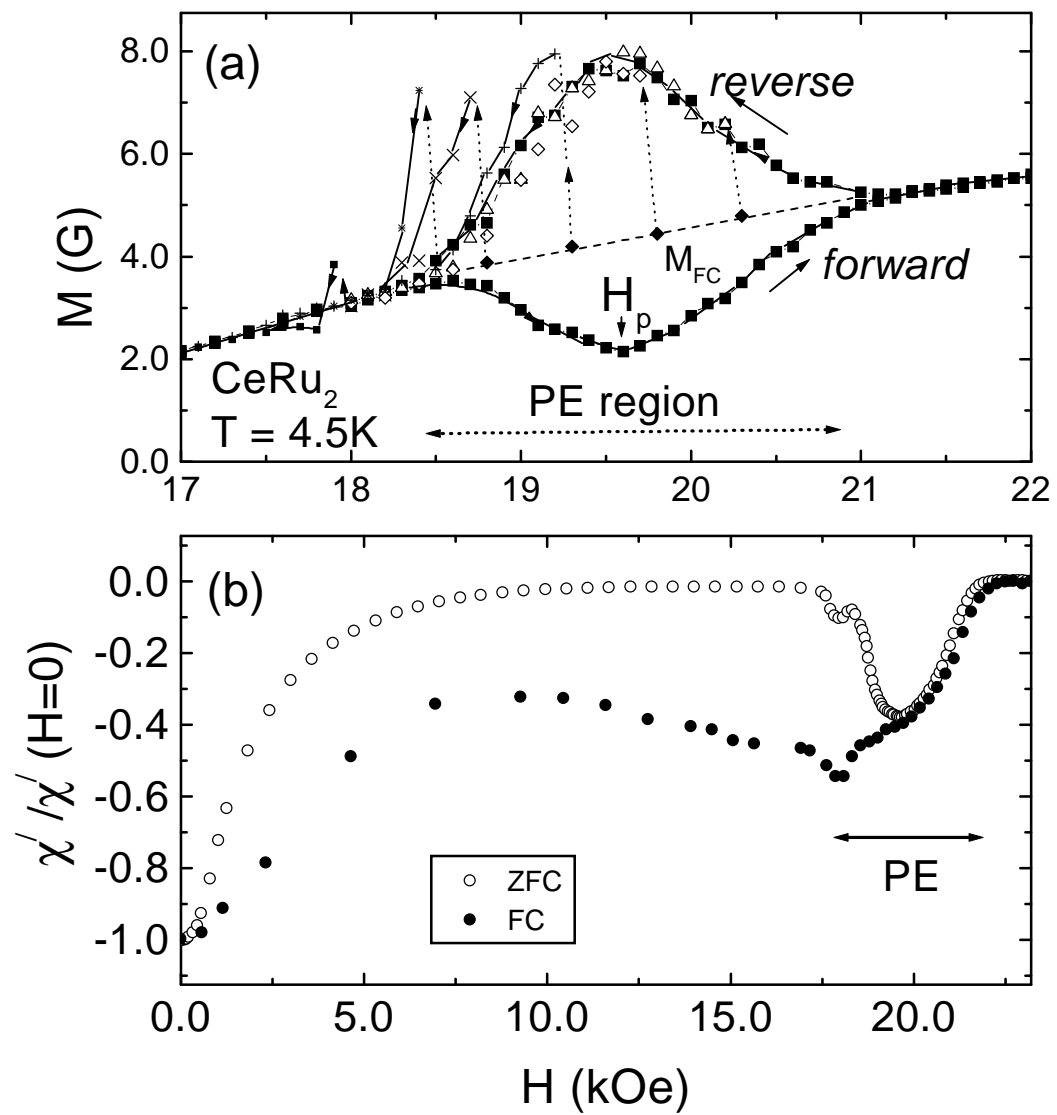
figure 1
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figure 2
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Figure 3
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