arXiv:cond-mat/9908222v1 [cond-mat.supr-con] 16 Aug 1999

A study of supercooling of the disordered vortex phase via minor hysteresis loops in 2H-NbSe$_2$

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We report on the observation of novel features in the minor hysteresis loops in a clean crystal of $NbSe_2$ which displays a peak effect. The observed behavior can be explained in terms of a supercooling of the disordered vortex phase while cooling the superconductor in a field. Also, the extent of spatial order in a flux line lattice formed in ascending fields is different from (and larger than) that in the descending fields below the peak position of the peak effect; this is attributed to unequal degree of annealing of the state induced by a change of field in the two cases.

PACS numbers :64.70 Dv, 74.25 Ha 74.60 Ge, 74.60 Jg

I. INTRODUCTION

A variety of recent transport, magnetic and structural studies in weakly pinned superconductor 2H-NbSe$_2$ support the view that the peak effect (PE) phenomenon [1] in the critical current density (J_c) marks an amorphisation of the flux line lattice (FLL) [2-10]. In a varying field experiment, for example, [6,9], $J_c(H)$ begins to increase from an initial low value at the onset field H_{pl}^+, reaches a maximum at the peak field H_p and eventually collapses below the measurable limit above the irreversibility field H_{irr} ($< H_{c2}$). The vortex matter is thought to undergo a transformation from a state with a high spatial order for $H < H_{pl}^+$ to a highly disordered state for $H > H_p$. This interpretation is usually rationalized within the Larkin-Ovchinnikov (LO) collective pinning formalism [11], where the Larkin volume V_c ($\approx R_c^2\ L_c$, where R_c and L_c are the radial and longitudinal correlation lengths, respectively) is a measure of the spatial extent of order in FLL and the critical current density is determined through the relation, $\mu_0 H J_c(H) \approx (n_p < f^2 > /V_c)^{1/2}$, where n_p and f are pin density and the elementary pinning force parameter, respectively. J_c measurements therefore can reveal the extent of spatial order in the FLL. Magnetization hysteresis measurements are a convenient tool for estimating $J_c(H)$ and thus for detecting the occurrence of phase transformations and associated changes in the vortex correlations across the PE regime.

Recently, the PE phenomenon has received a great deal of attention due to a characteristically rich phenomenology that accompanies it. Prominent among them is the marked history dependence in the critical current. In the LO scenario, this translates into a history dependence of the Larkin volume V_c, since quantities such as f and n_p cannot be history dependent. The history dependence of the correlations implies strong metastability and is thus a hallmark of disorder in condensed matter systems. Recent theoretical studies emphasizing the role of quenched random disorder in producing novel disordered (glassy) phases (see, for instance, Refs. 12-14) further illustrate the need to understand and ultimately unravel the complex effects of disorder readily observed in the PE regime. Ravikumar et al [9] have experimentally demonstrated via dc magnetization technique the presence of a highly disordered vortex state when a sample is field cooled (FC) in $H < H_p$[6]. In crystals of $2H\text{-}NbSe_2$ and $CeRu_2$ with comparable levels of effective pinning, they had shown that the critical current density in the FC state is larger than that in the zero field cooled (ZFC) state for $H < H_p$, i.e., $J_c^{FC}(H) > J_c^{ZFC}(H)$. They had also shown that the disordered FC state could be annealed to the ordered ZFC like state when the sample was subjected to a small change in magnetic field. Independently, a different type of history effects has been reported [15,16] in a wide variety of polycrystalline and single crystal samples of pure and doped $CeRu_2$ showing PE phenomenon. In these cases, the minor magnetization curves, starting at a field H on the forward branch of the hysteresis loop (such that $H_{pl}^+ < H < H_p$), were reported to saturate without merging with the reverse leg of the hysteresis loop. The minor curves starting on the reverse branch, on the other hand, merge with the forward branch of the hysteresis loop. Roy et al [15] attribute the observed behavior to an *anomalous* nature of PE phenomenon in the mixed valent superconducting system $CeRu_2$, in contrast with, and as distinct from, the conventional PE in $2H\text{-}NbSe_2$[2] and most other weakly pinned low T_c superconductors [9,10,17]. They propose [15,16] that this novel behavior of the minor loops reflects a thermodynamic evidence for a first order transformation to a new phase caused by the positional dependence of the order parameter in superconductors with large normal state paramagnetism, like, heavy fermion superconductors, mixed valent rare earth systems, etc. [18]. This is in contrast to the explanation based on metastability effects in the vortex matter caused by quenched disorder.

Very recently, a similar effect with the minor hysteresis curves has been observed in a cuprate superconductor as well [19]. The microscopics in the cuprate system appears to bear little resemblance to that in the mixed valent systems such as $CeRu_2$. We propose that the above sets of experiments [9,15,16,19] exemplify the ubiquitous nature of thermomagnetic history dependence of J_c in superconductors in general and thus require an explanation that is independent of the microscopics relevant for superconductivity in the diverse systems in which these effects are present.

In this paper, through detailed measurements of minor magnetisation curves on a clean single crystal of $2H\text{-}NbSe_2$ (belonging to the category of most weakly pinned samples of type II superconductors [2,4]), we present an understanding, based on the LO collective pinning description [11] for the observed path dependence in the critical currents and vortex correlations. We invoke the notion of a disorder-assisted supercooling of a metastable disordered phase, that is otherwise thermodynamically stable only above H_p. We further propose that changes in the applied field act as a driving force that helps to anneal the system, often partially. As a result, systems with different field histories are often in metastable states with different degrees of annealing and thus with different values of vortex correlations, leading to the different critical currents, as observed experimentally. In the present work, three types of isothermal minor hysteresis loops were studied : (I) Decreasing field after cooling the sample in a field (FC-REV), (II) and (III) : decreasing /increasing field from a given point on the forward/reverse leg of the envelope hysteresis curve. Fig.1 shows a schematic view of all these loops for the case where $J_c(H,T)$ is single-valued, independent of the field/temperature history. In all such cases, the minor curves merge into the envelope curve while always remaining within it, without any overshoot effects. We show below that violations of this standard scenario provide an understanding of the aforementioned anomalous behavior.

II. EXPERIMENTAL

We carried out *dc* magnetization measurements using a Quantum Design (QD) SQUID magnetometer (Model MPMS5) on a $2H\text{-}NbSe_2$ single crystal with $T_c \approx 7.25$ K. The crystal was mounted with field applied parallel to its c-axis. The thermomagnetic history dependent measurements have been performed at 6.95 K, where a well recognized peak effect, manifesting as a sharp increase in the magnetization hysteresis, is observed at a relatively low field of about 1000 Oe. Occurrence of PE at such low fields, where the flux line lattice constant a_0 (= 1600 A) is of the same order as the range of interaction (i.e., the penetration depth λ_{ab}) at 6.95K in this system [20], confirms that the sample has weak quenched disorder [7]. In the field range $1-2$ kOe, the inhomogeneity experienced by the sample in a 2 cm full-scan in a QD SQUID magnetometer is of the order of 0.1 Oe[21]. This value is much smaller than the threshold field H_{II} [22] required to change the sign of the induced shielding currents throughout the sample. H_{II} value is estimated by measuring the minor magnetisation curves [23,24] for fields above H_p and was found to be about 10 Oe. Thus, the field inhomogeneity in a full scan of 2 cm does not introduce any error in the measured magnetization values, and we have recorded all the present data with a 2 cm scan length instead of *half scan technique* utilized earlier by Ravikumar et al [9,21], in their study of $CeRu_2$ and more strongly pinned sample of 2H-$NbSe_2$.

III. RESULTS AND DISCUSSION

In Fig.2, we show the magnetization hysteresis loop in the present 2H-$NbSe_2$ crystal at 6.95 K, indicating the onset field of PE on the ascending field cycle ($H_{pl}^+ \approx 800$ Oe), the peak field H_p (≈ 1000 Oe) and the irreversibility field H_{irr} (≈ 1250 Oe). Within the LO collective pinning description, the Larkin volume V_c begins to shrink at H_{pl}^+ and FLL reaches nearly amorphous (though pinned) state at H_p [7,10]. Note that the onset of PE is much sharper (see inset(ii)) on the ascending field cycle than on the descending field cycle. This immediately shows that at a given field value in the PE region, J_c and thus V_c in the vortex state are not the same on the ascending and descending field cycles in the PE regime. In what follows, we examine this non-uniqueness of the vortex correlations in greater detail.

In Fig.3(a), we show the magnetization curves measured for FC-REV case, i.e., magnetization of a field cooled sample measured in decreasing magnetic fields. The field value in which the sample was cooled was varied across the PE region. Note that in Fig.3(a), the FC-REV curve originating at $H > H_p$ merges with the reverse magnetization envelope curve [23,24] in accordance with the critical state model. On the other hand, FC-REV curves, originating from a field $H < H_p$, overshoot the reverse magnetization envelope curve in a clear departure from the conventional behavior shown in Fig.1. The difference between the highest magnetisation value recorded on the FC-REV curve and the notional equilibrium magnetisation value could be taken as a measure of the critical current density in the FC state [25]. Thus, the minor curves in the FC-REV case produce higher J_c values than those in the conventional descending part of the envelope curve.

This anomalous behavior can be explained by assuming a supercooling of the disordered phase and the annealing effect (i.e., increase in vortex lattice correlations or growth of Larkin domain volume V_c) due to a subsequent field change. In the FC state, the FLL traverses through a pinned amorphous state as it is cooled down across the $H_p(T)$ line (see the phase diagram drawn in the inset (i) of Fig.2). The highly disordered vortex state that is stable above PE curve, with a large density of defects/dislocations [2,5,7], is then effectively supercooled when the sample is cooled to a given T in a field less than H_p(T). Vortex state obtained by cooling the sample in a field H is more disordered than that at the same field value on the descending branch of the envelope loop. In the latter case, the process of lowering of field (below H_p) induces partial annealing and produces a vortex state, which is more correlated and thus has a smaller critical current than that in the field cooled state. Furthermore, annealing induced by the field change as mentioned above is also clearly seen in the FC-REV magnetization values moving towards the reverse envelope curve (see inset of Fig.3(a)).

The FC-REV curves initiated from different fields below H_p form a family of curves. FC-REV curve originating from $H = H_p$ essentially retraces the reverse magnetisation envelope curve below H_p. As stated earlier, the vortex state at any field value below H_p on the reverse magnetization curve is the result of a gradual healing of the disordered state existing at $H = H_p$. Thus, the reverse magnetization envelope curve (for $H < H_p$), in principle need not, and in practice will not, be a mirror reflection of the forward magnetization envelope curve as the specific kinetics of the annealing processes are different. That the process of healing of FLL dislocations could continue down to a field well below H_{pl}^+ (during the descending cycle) is a clear indicator of this difference. The data shows that for a given field $H(< H_p)$, the Larkin domains on the reverse magnetization envelope curve are smaller than those on the forward magnetization envelope curve. In other words, J_c values on the descending field cycle are larger than those on the ascending field cycle. This difference is likely to be due to the difference in the starting configurations on the ascending and descending branches. For the latter, one starts from a much more disordered state; thus the residual disorder after a comparable level of (incomplete) annealing is more than that in the former.

Using the scenario described above, we now examine the minor magnetisation curves of the type II and III (cf. Fig.1) in the PE region of $2H{-}NbSe_2$ and compare and contrast them with anomalous behavior of minor magnetisation curves in the PE region of $CeRu_2$ [15,16]. In Fig.3(b), we first show the minor magnetization curves of type II, initiating from different fields lying on the forward magnetization curve. The minor magnetization curves initiated from $H > H_p$ and $H < H_{pl}^-$ merge with the reverse magnetization curve within a field change of about 10 Oe. The threshold field H_{II} is thus estimated from these curves to be of the order of 10 Oe. However, for $H_{pl}^- < H < H_p$, the minor magnetization curves do not merge into the reverse envelope curve, because $J_c(H)$ values on the ascending field cycle are smaller than those on the descending field cycle, as asserted earlier. The significance of H_{pl}^- is that the disorder present at $H = H_p$ is maximally annealed at this field value on the descending cycle.

In Fig. 3(c), we show the measured minor magnetization curves of the type III, i.e., by increasing the field from different points on the reverse magnetization envelope curve. These minor curves now overshoot the forward envelope magnetisation curve when the field is increased by about 10 Oe. The annealing due to the field increase (of about 10 Oe) is inadequate to produce a comparable level of lattice order existing at the corresponding fields on the forward envelope loop. When the field is further increased, the residual disorder gets annealed. Thus the minor magnetisation curves eventually merge into the more ordered forward envelope curve. However, Roy et al[15] reported that in $CeRu_2$ the minor curves initiated from the reverse magnetisation envelope curve readily merge with the forward magnetisation curve, in contrast with our data. Whether this is a significant difference, or merely a trivial one, in the comparative levels of annealing in the two instances, is unclear [26].

The critical current densities, J_c^{for} and J_c^{rev} on the ascending and descending field branches can now be estimated from the maximum width of these two sets of minor magnetisation curves. We collate, in Fig.4, the relative $J_c(H)$ values corresponding to three thermomagnetic histories of the sample, viz., the FC state and the states along the *forward* and the *reverse* legs of the envelope hysteresis loop. The three sets of $J_c(H)$ values have been estimated [9,15,25] by taking the notional half width of the magnetization hysteresis at a given H to be proportional to the corresponding critical current density. The data in Fig.4 can be summarized by the inequality,

$$J_c^{FC}(H) > J_c^{rev}(H) > J_c^{for}(H),$$

which is consistent with an early report of transport data in a Nb crystal by Steingart et al [27]. In the framework mentioned above, this inequality corresponds to the least annealed state in the FC mode and the most annealed state on the forward curve, while the J_c^{rev} is intermediate between the two.

IV. CONCLUSION

We propose that the PE phenomenon marks a true thermodynamic phase transformation between an ordered solid that is stable below H_{pl}^+ (on forward envelope loop) and a fully disordered vortex state that is stable above H_p. Further, this transformation is first order in character. Thus, it is possible to *supercool* or *superheat* one phase into the regime of stability of the other phase. For the PE regime, when the free energies of the ordered and disordered phases are not significantly different, the metastability effects are expected to be prominent. Moreover, thermal fluctuations are inadequate, at least below H_p, for the system to fully explore the phase space, which helps the metastability of the *supercooled* phase, aided by the fact that both phases have finite pinning. Substantial driving forces experienced when the field is changed allow the system to anneal (or fracture, as the case may be) towards the stable state. When the levels of annealing are incomplete and, as is often the case, unequal due to the previous thermomagnetic history, the system will typically exhibit different levels of correlations and thus different critical currents, as expected within the LO mechanism. The history dependence of critical current should thus be a generic occurrence in this regime for comparable levels of disorder. We emphasize that the proposed explanation of the various anomalous history dependent magnetization hysteresis data shown here and elsewhere [9,15,16,19] are in terms of an order-disorder transformation and disorder-aided *supercooling*. This explanation is , prima facie, independent of the specifics of microscopic considerations consistent with the ubiquitous nature of the phenomena under consideration. It is tempting to suggest that the so-called *anomalous* PE in $CeRu_2$ [15,16,18] may also find an explanation within the scenario described above; whether this is indeed the case remains to be concluded.

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26. Inspection of Fig.3 in Ref.15 (J. Phys : Condens. Matter 10, 4885(1998)) suggests that the minor loop initiated from the reverse leg indeed overshoots the forward envelope curve even in $CeRu_2$.

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FIGURE CAPTIONS

Fig. 1. Schematic behavior of minor magnetisation curves initiated from field cooled (I) magnetisation value (M_{FC}) and those from forward (II) and reverse (III) legs of the envelope hysteresis loop within the framework of critical state model, i.e., assuming J_c(H) is uniquely defined by H. The field interval H_{II}, corresponding to a threshold field change required to change the sign of shielding currents throughout the sample, is also indicated.

Fig.2. Magnetization hysteresis of a $NbSe_2$ crystal recorded at 6.95K for $H \parallel c$. The inset (i) schematically shows three different paths, viz., the zero field cooled (ZFC), field cooled (FC) and descending fields from above H_{c2}. The PE line H_p(T) and the upper critical field line H_{c2}(T) have been determined from the temperature dependent in-phase ac susceptibility data, as in Ref.3. Note that in the FC mode, the sample would cross the $H_p(T)$ line at different point each time, while reaching a given (H, T) value. Inset (ii) shows an enlarged view of the M-H loop in the PE region, indicating the onset field H_{pl}^{+}, the peak field H_p and the irreversibility field H_{irr}.

Figs.3(a) to 3(c). Minor magnetisation curves in the given $NbSe_2$ crystal at 6.95 K measured along three paths, as schematically sketched in Fig.1. The inset in Fig.3(a) shows the merger of FC-REV curves initiated from M_{FC}(H) (where $H = 0.4$ kOe and 0.6 kOe) into the reverse envelope curve. Note that the minor curves in Fig.3(b) do not readily overlap with reverse envelope curve, whereas those in Fig.3(c) cut across the forward envelope curve.

Fig.4. Field dependence of J_c for $H \parallel c$at 6.95K in $NbSe_2$ for three different histories as indicated.

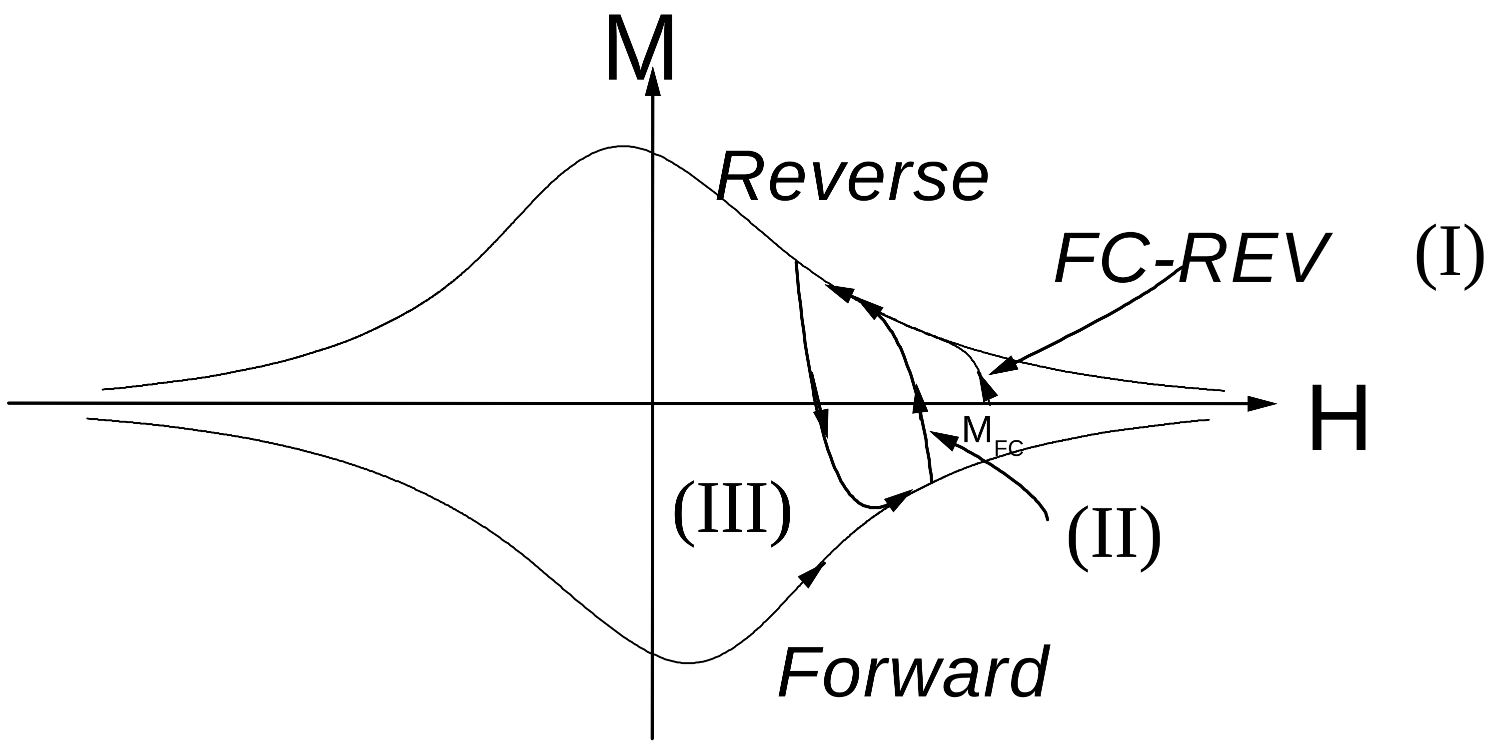
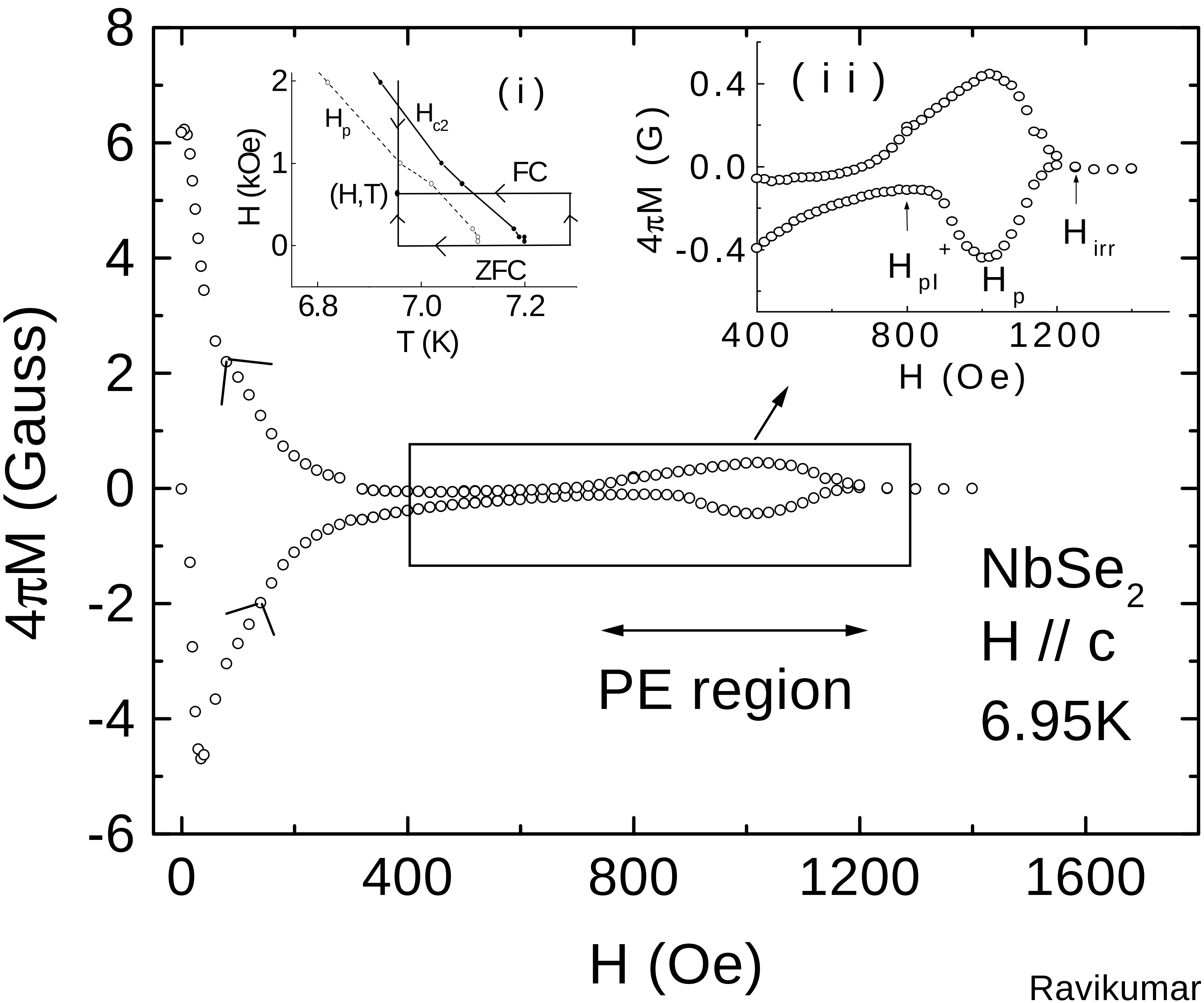
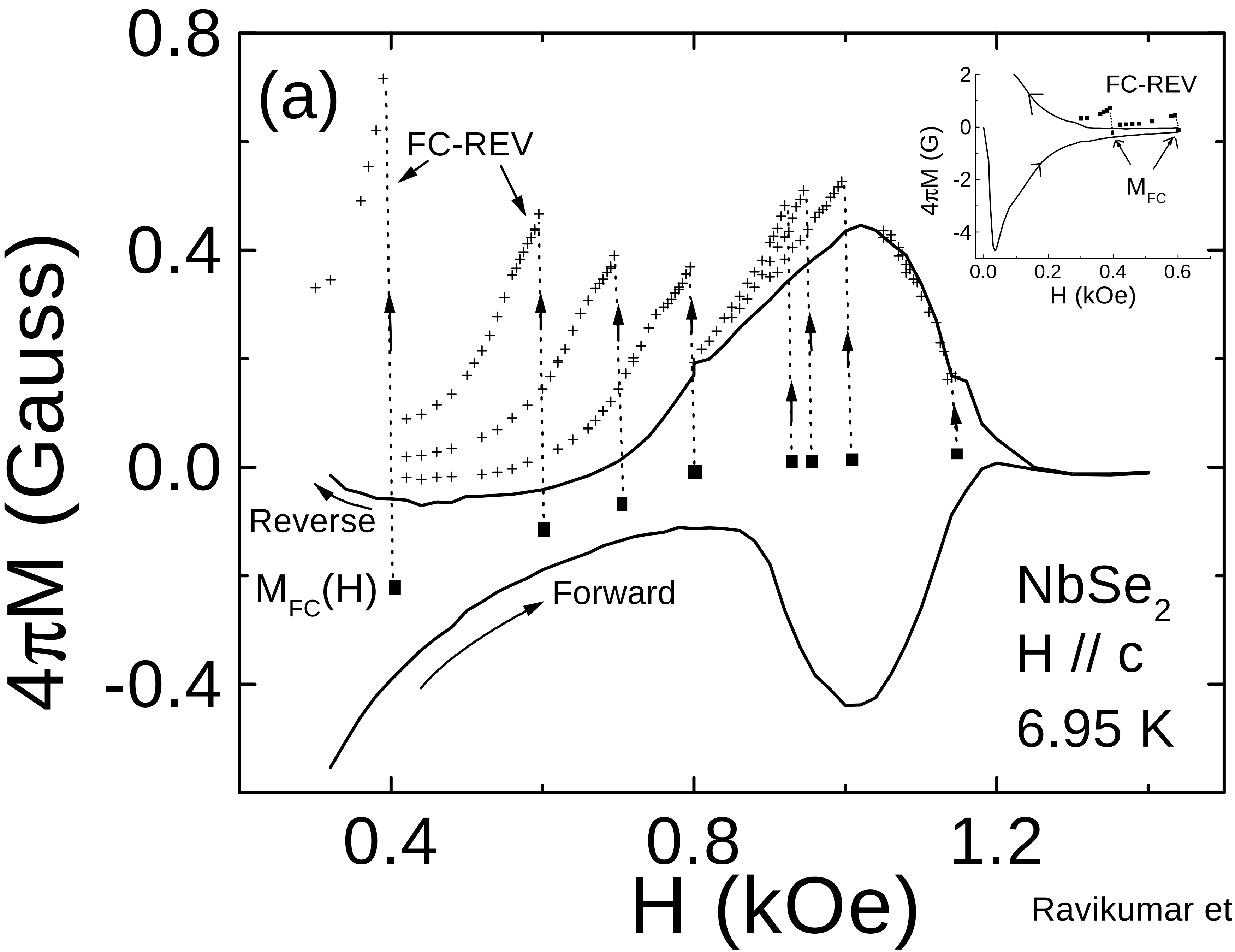


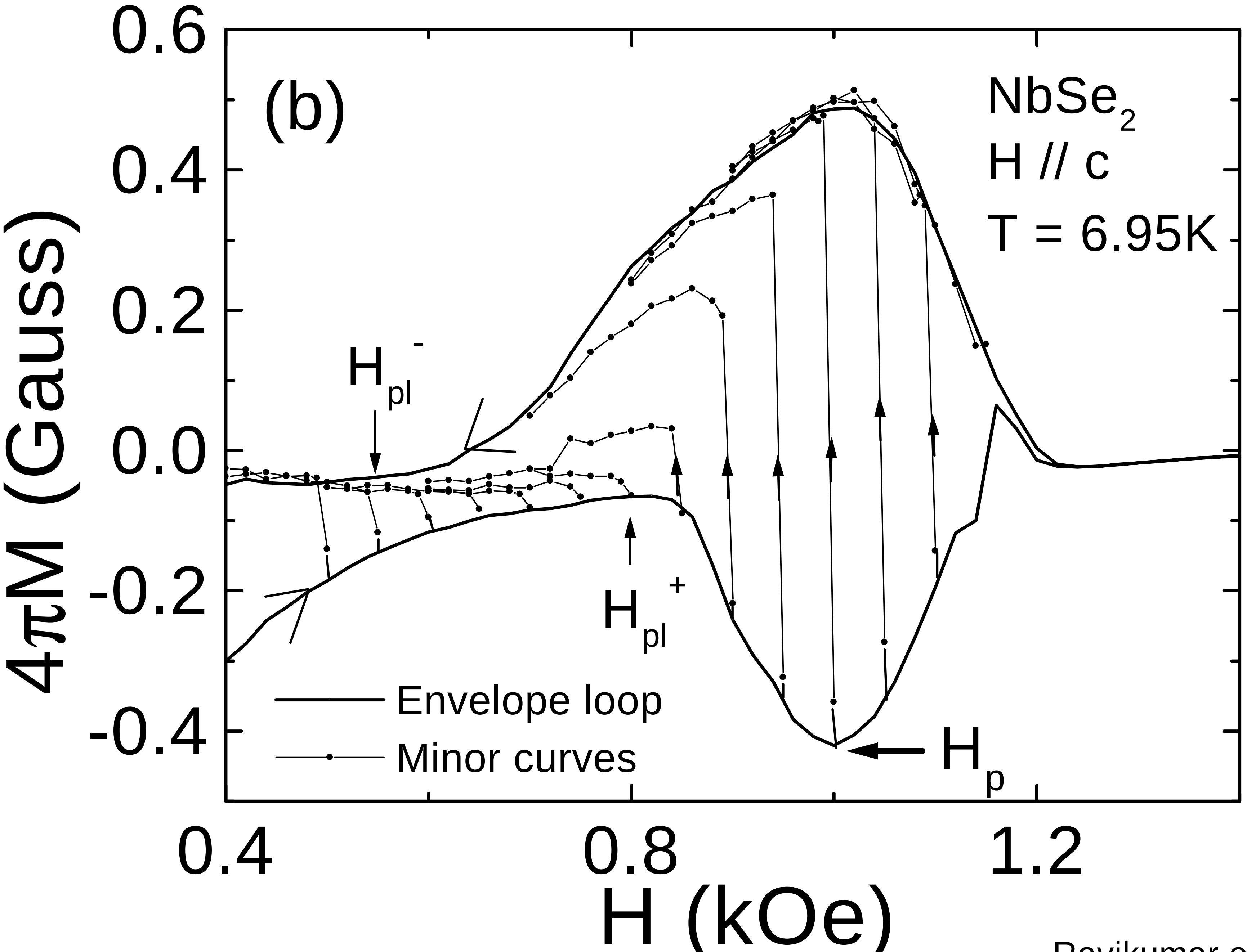
Fig.1a -Ravikumar et al



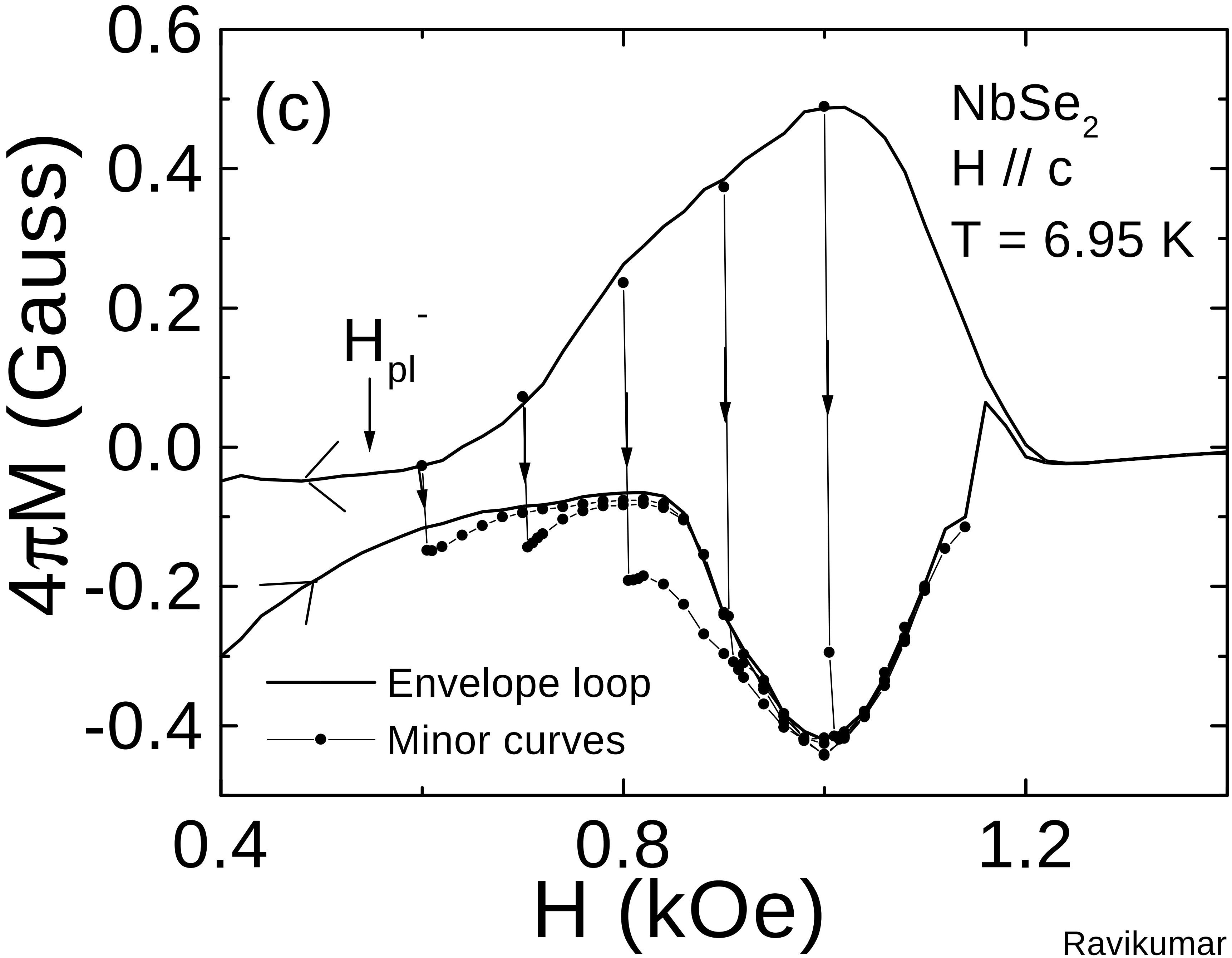
Ravikumar et al- fig.2



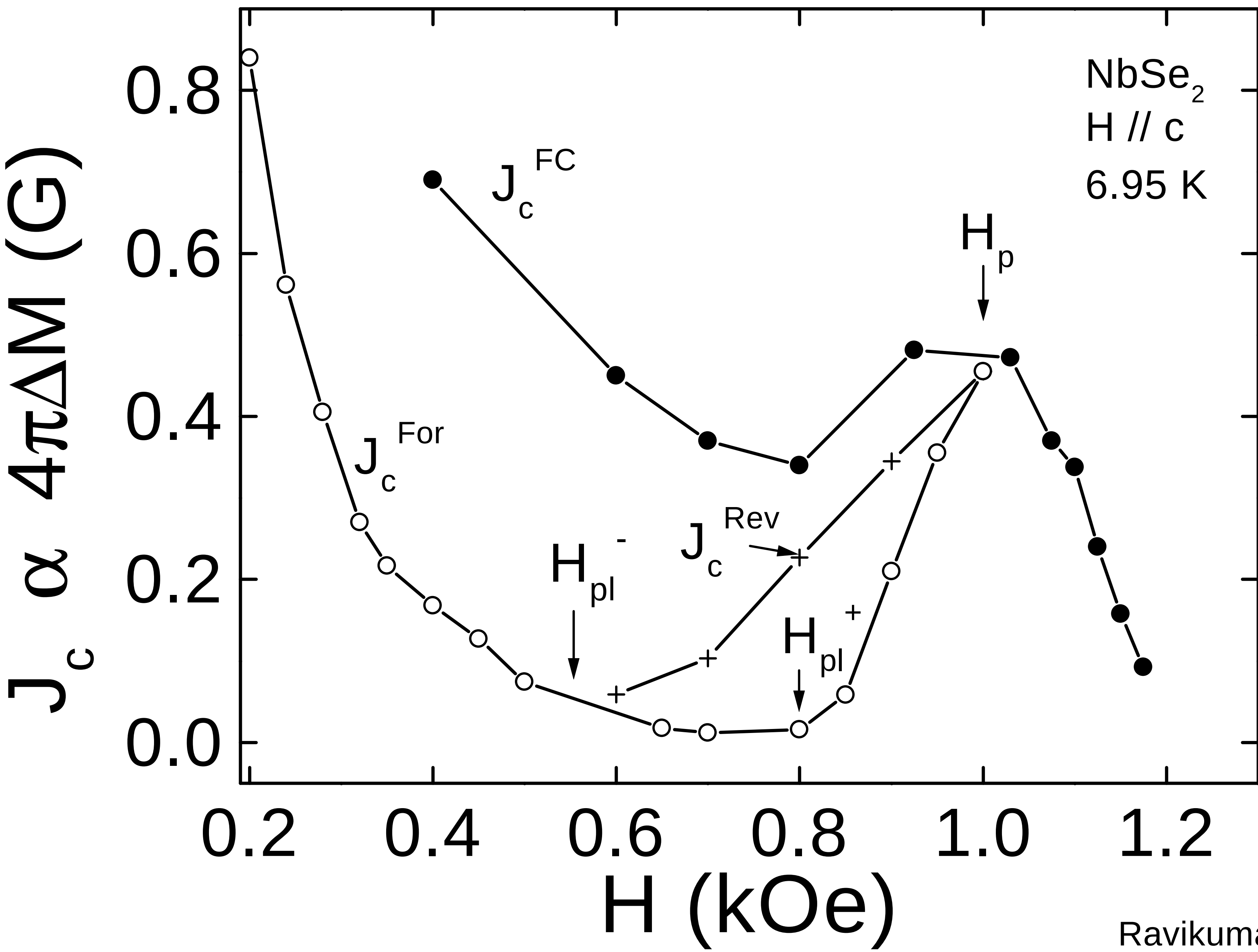
Ravikumar et al - fig.3a



Ravikumar et al - fig.3b



Ravikumar et al - fig.3c



Ravikumar et al Fig.4