A study of supercooling of the disordered vortex phase via minor hysteresis loops in 2H-NbSe₂

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We report on the observation of novel features in the minor hysteresis loops in a clean crystal of NbSe₂ 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-NbSe₂ 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_c[2]). 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 (≈ R_c² 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, μ0H J_c(H) ≈ (n_p < f² > /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 its 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-NbSe₂ and CeRu₂ 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_cFC(H) > J_cZFC(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 CeRu₂ 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 CeRu₂, in contrast with, and as distinct from, the conventional PE in 2H-NbSe₂[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-$NbSe_2$ (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-$NbSe_2$ 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 $\lambda_{ab}$) 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 $H_{11}$ [22] required to change the sign of the induced shielding currents throughout the sample. $H_{11}$ 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 $H_p$ ($\approx 1000$ Oe) and the irreversibility field $H_{irr}^+$ ($\approx 1250$ 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. In other words, smaller than those on the forward magnetization envelope curve are the minor magnetization curves in the PE region of \(2\text{H}^{\text{II}}\) (Fig.1) in the PE region of \(2\text{H}^{\text{II}}\) as contrasted with anomalous behavior of minor magnetization curves. We collate, in Fig.4, the data in Fig.4 can be summarized by the inequality,

\[
J_c^{\text{FC}}(H) > J_c^{\text{rev}}(H) > J_c^{\text{for}}(H),
\]

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 \(J_c^{\text{rev}}\) 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...
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 \( \text{CeRu}_2 \) \([15,16,18]\) may also find an explanation within the scenario described above; whether this is indeed the case remains to be concluded.

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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 $H_{on}^p$, 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. 3a

(a) $4\pi M (\text{Gauss})$

$M_{FC}(H)$

$H (\text{kOe})$

$H / / c$

$6.95 \text{ K}$

$\text{NbSe}_2$
\( \mathbf{H} \) (kOe)

\( 4\pi M \) (Gauss)

\( H_{pl}^- \)

\( H_{pl}^+ \)

\( H_p \)

\( \text{NbSe}_2 \)

\( H // c \)

\( T = 6.95 \text{K} \)

Envelope loop

Minor curves

Ravikumar et al - fig.3b
Figure 3c: 

- NbSe$_2$
- $H \parallel c$
- $T = 6.95$ K

Graph showing the magnetic moment $4\pi M$ (Gauss) against magnetic field $H$ (kOe) with envelope loop and minor curves indicated. 

Ravikumar et al - fig.3c
