Stable and metastable vortex states and the first-order transition across the peak-effect region in weakly pinned $2H$-$NbSe_2$

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The peak effect in weakly pinned superconductors is accompanied by metastable vortex states. Each metastable vortex configuration is characterized by a different critical current density J_c, which mainly depends on the past thermomagnetic history of the superconductor. A recent model [G. Ravikumar *et al.*, Phys. Rev. B **61**, R6479 (2000)] proposed to explain the history-dependent J_c, postulates a stable state of vortex lattice with a critical current density J_c^{st} determined uniquely by the field and temperature. In this paper, we present evidence for the existence of the stable state of the vortex lattice in the peak-effect region of $2H$-$NbSe_2$. It is shown that this stable state can be reached from any metastable vortex state by cycling the applied field by a small amplitude. The minor magnetization loops obtained by repeated field cycling allow us to determine the pinning and ''equilibrium'' properties of the stable state of the vortex lattice at a given field and temperature unambiguously. The data imply the occurrence of a first-order phase transition from an ordered phase to a disordered vortex phase across the peak effect.

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I. INTRODUCTION

In the presence of strong pinning, the vortex state of type-II superconductors is usually characterized by the critical current density $J_c(H,T)$ that decreases monotonically with increasing field H or temperature T. In weakly pinned superconductors, on the other hand, the interplay between intervortex interaction and flux pinning produces an anomalous peak in J_c as a function of both field and temperature[1] just below the normal-state boundary [usually designated as the peak effect (PE)]. Within the collective pinning description,[2] this signifies the vortex phase undergoing a transition/crossover from an ordered state to a disordered state.[1,3–5] The detailed nature of this transition, e.g., whether it is a thermodynamic phase transition or not, remains a subject of considerable debate.

One of the key issues is the detection of an anomaly in the thermodynamic quantities such as specific heat or equilibrium magnetization M_{eq}. J_c and M_{eq} can be estimated from the measured irreversible magnetization data of a superconducting sample[6–8] using the relations

$$J_c(H)=[M(H\downarrow)-M(H\uparrow)]/2g\mu_0 R, \tag{1a}$$

$$M_{eq}(H)=[M(H\uparrow)+M(H\downarrow)]/2, \tag{1b}$$

where $M(H\uparrow)$ and $M(H\downarrow)$ are the magnetizations in the increasing (forward) and decreasing (reverse) field cycles, respectively, $\mu_0=4\pi\times10^{-7}$ W/A m, R is the sample dimension transverse to the applied field, and g is a factor depending on the sample geometry. Equation (1) implicitly assumes that J_c is history independent and is thus uniquely determined by the local induction B. However, across the peak-effect region, the above equations are not valid due to a strong history dependence in J_c.[9–17]

Recently, considerable effort has gone into ascertaining the equilibrium magnetization across the peak-effect region where an order–disorder transition occurs in the vortex matter. However, these efforts have met with ambiguous and somewhat conflicting results. For example, the construction of the equilibrium magnetization from the hysteresis loop by using two different kinds of minor magnetization curves[16,18,19] results in apparently different conclusions. In one case, a jump[16,19] in M_{eq} could be found at the onset of PE, while, in the other case, there may be no increase at all.[18] These differences apparently originate from the difficulties in establishing an unambiguous and reproducible vortex state due to a strongly history-dependent metastability in the PE region. The different procedures proposed to obtain M_{eq} will be discussed in Sec. II.

In Sec. III, we briefly discuss a recent phenomenological model[20] that addresses the issue of the history-dependent J_c

and the metastability in the vortex state through an extension of the Bean's critical state model.[6] In Sec. IV, we present an experimental method based on the ideas of the model[20] to obtain a unique ''stable'' vortex state in the PE region that is independent of the past magnetic history. We propose that this state, in effect, is the stable or equilibrium state and evaluate the critical current density and M_{eq} of this state. We further demonstrate that a sharp change in the equilibrium magnetization (albeit smeared) occurs across the PE region. These results imply that an underlying first-order phase transition, presumably driven by a competition between elastic and pinning energies in a situation where thermal fluctuations are weak, marks the peak effect.

II. MINOR CURVES AND THE EQUILIBRIUM MAGNETIZATION ACROSS THE PEAK EFFECT

In the peak-effect region, the critical current density in the increasing field cycle $J_c(H\uparrow)$ is less than that $[J_c(H\downarrow)]$ in the decreasing field cycle[9,10,14] for $H<H_p$, where H_p is the field where J_c is maximum. However, well below the onset of the PE and at $H>H_p$, J_c is independent of the magnetic history. One of its consequences is the peculiar behavior of the minor magnetization curves, which cannot be reconciled within the critical state model.[6] For instance, a typical minor magnetization curve (type I) initiated from a field $H<H_p$ (on the forward magnetization curve) in the PE region *saturates without meeting* the reverse magnetization curve,[14,16,17] as shown in Fig. 1(a). On the other hand, the minor curves (type II) measured by increasing the field from different points on the reverse magnetization curve *overshoot* the forward curve,[14,18] as shown in Fig. 1(b). However, this anomalous behavior contrasts with the conventional behavior for both types of the minor curves starting at (a) $H>H_p$ and (b) well below the PE region, which meet the magnetization envelope, constituted by the forward and reverse curves, as expected from the Bean's critical state model.

A new procedure was proposed[16] to obtain M_{eq} from the minor magnetization curves of type I by the relation

$$M_{eq}(H)=[M(H+\delta,\uparrow)+M_{ML}(H-\delta,\downarrow)]/2, \qquad (2)$$

where $M(H+\delta,\uparrow)$ is the magnetization at a field $H+\delta$ [denoted by point A in Fig. 1(a)], where the minor curve is initiated on the forward curve. $M_{ML}(H-\delta,\downarrow)$ is the magnetization on the minor curve at a field $H-\delta$, where it saturates as indicated by point B in Fig. 1(a). This procedure is based on the implicit assumption that the vortex state formed on the forward curve is an equilibrium state. This assumption is, however, inconsistent with the experimental observation by Wordenweber, Kes, and Tsuei,[10] who showed that both current cycling and field cycling processes eventually establish a vortex state with a J_c higher than that on the forward curve. Such an observation indicates that the vortex state formed on the forward curve is metastable in nature.

Tenya *et al.*[18] have preferred a procedure given below that is very similar to the one described above, but using the minor curves of type II described in Fig. 1(b):

$$M_{eq}(H)=[M(H-\delta,\downarrow)+M_{ML}(H+\delta,\uparrow)]/2, \qquad (3)$$

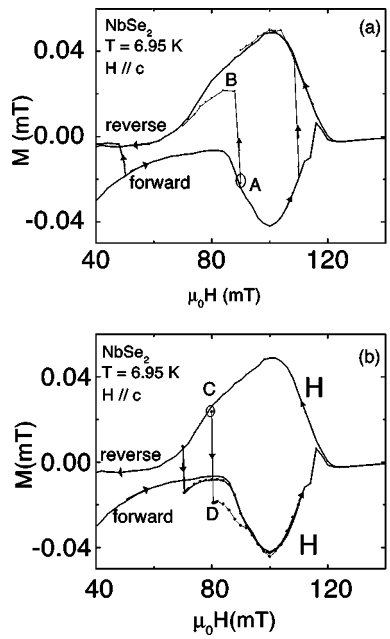

FIG. 1. Typical magnetization hysteresis loop observed in the peak-effect region of a superconducting $2H$-$NbSe_2$. In (a), minor curves (type I) obtained by decreasing the field from A, corresponding to a field $(H+\delta)$ on the forward magnetization curve is shown to saturate at B corresponding to a field $(H-\delta)$. Magnetization values at A and B are $M(H+\delta,\uparrow)$ and $M_{ML}(H-\delta,\downarrow)$, respectively (see text). In (b), minor curves (type II) obtained by increasing the field from C, corresponding to a field $(H-\delta)$ on the reverse magnetization curve that saturates at D $(H+\delta)$ corresponding to a magnetization value $M_{ML}(H+\delta,\uparrow)$.

where $H-\delta$ [point C in Fig. 1(b)] is the field where the minor curve is initiated on the reverse curve and $H+\delta$ [point D in Fig. 1(b)] is the field where it saturates. $M_{ML}(H+\delta,\uparrow)$ is the saturated magnetization value on the minor curve. This procedure, too, has the shortcoming similar to that in Eq. (2), viz., the vortex state on the reverse magnetization curve is actually a metastable state.[10,14,16] Moreover, not only are these recipes deficient, they also yield different conclusions, viz., an enhancement in equilibrium magnetization is observed in one case, whereas it is absent in the other. These ambiguities point to the need to evolve a more satisfactory procedure to arrive at a unique and stable vortex state unambiguously and determine the equilibrium magnetization assuming the stable state to be the equilibrium state.

III. MODEL FOR HISTORY EFFECTS AND METASTABILITY

Ravikumar *et al.*[20] incorporated the history dependence in the macroscopic critical current density J_c by postulating

$$J_c(B+\Delta B)=J_c(B)+(|\Delta B|/B_r)(J_c^{st}-J_c), \qquad (4)$$

where the critical current density $J_c(B)$ is a macroscopic representation of a particular metastable configuration of the vortex lattice at a field B. Equation (4) describes how the vortex state evolves from one metastable configuration to another. An important assumption of this model is the existence of a stable vortex state with a critical current density J_c^{st}, which is unique for a given field and temperature. B_r is a macroscopic measure of metastability and describes how strongly J_c could be history dependent. In the limit of B_r tending to zero, however, this model reduces to the standard critical state model for which J_c $(=J_c^{st})$ is independent of the magnetic history. It can be seen from Eq. (4) that a metastable vortex state with $J_c \neq J_c^{st}$ can be driven into a stable state by merely oscillating the field by a small amplitude (see Fig. 1 of Ref. 20). In the PE region, the energy barriers between different metastable vortex configurations are much greater than the available thermal energy. The field cycling allows the vortices to move and explore the energy landscape and thereby rearrange into a vortex configuration closer to the stable state. In the next section, we will demonstrate this experimentally and show that the stable state obtained is indeed independent of the magnetic history.

In the limit $\Delta B \rightarrow 0$, Eq. (4) can be rewritten in the form

$$\pm dJ_c/dB = (J_c^{st} - J_c)/B_r, \tag{5}$$

where upper and lower signs are applicable in the cases of increasing and decreasing local field B, respectively. In each case $J_c(B)$ can be obtained by solving Eq. (5), provided the functional forms of $J_c^{st}(B)$ and $B_r(B)$ are known. We assume for $J_c^{st}(B)$ and $B_r(B)$ the following forms, used in Ref. 20 for calculating the minor magnetization curves:

$$J_c^{st}(B) = J_{c1}(1 - B/\mu_0 H_1) + J_{c2} e^{-(B-\mu_0 H_p)^2/2\mu_0 H_W^2} \tag{6}$$

and

$$B_r(B) \approx (B - \mu_0 H_{low})^m (\mu_0 H_p - B)^n \text{ for } H_{low} < B/\mu_0 < H_p$$
$$\approx 0 \quad \text{otherwise.} \tag{7}$$

The first term in Eq. (6) is the field dependence of J_c^{st} well below the peak, and the second term reflects the peak in J_c^{st} vs B. $B_r(B)$ in Eq. (7) accounts for the observed history dependence in J_c in the PE region. $B_r = 0$ in the field ranges $H < H_{low}$ and $H > H_p$ signifies that J_c is independent of magnetic history and is always equal to J_c^{st}. For the two limiting cases $H < H_{low}$ and $H > H_p$, intervortex interaction and flux pinning are dominant, respectively, and therefore the stable state is readily accessed by the vortex lattice. The values of different parameters used in this paper are listed in the caption of Fig. 2. $J_c(H\uparrow)$ $[J_c(H\downarrow)]$ is calculated by numerically solving Eq. 5 with an upper (lower) sign with the initial condition $J_c(H\uparrow)$ $[J_c(H\downarrow)] = J_c^{st}(H)$ at some field below H_{low} (above H_p). In Fig. 2(a), we present an evaluation of $J_c(H\uparrow)$ and $J_c(H\downarrow)$ that obey the inequality $J_c(H\uparrow) < J_c^{st}(B) < J_c(H\downarrow)$. It was earlier interpreted that the vortex state formed on the decreasing field cycle is a *supercooled* disordered state.[14] In other words, the vortex state formed in the decreasing field (from above H_p) retains the memory of the vortex correlations from the previous fields. In analogy, we can argue that the vortex state formed on the increasing field cycle is a *superheated* ordered state. Both of these states are metastable in nature. As argued above, they can be driven into a stable state by oscillating the external field by a small amplitude.

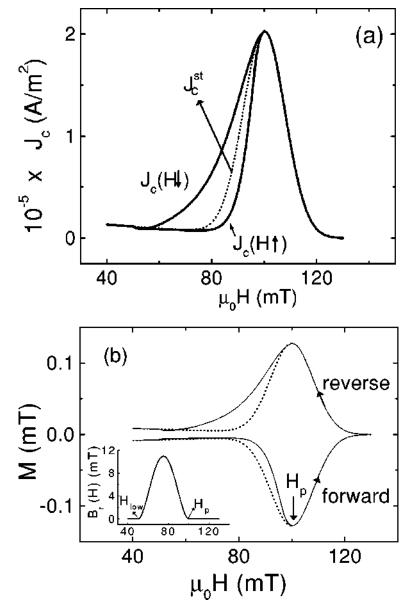

FIG. 2. (a) Calculated critical current densities $J_c(H\uparrow)$ and $J_c(H\downarrow)$ in the increasing and decreasing field cases, respectively. These are compared with the stable critical current density J_c^{st} (dotted line). In this calculation we used $H_{low} = 0.05$ T, $H_p = 0.1$ T, $J_{c1} = 10^4$ A/m^2, $J_{c2} = 20 J_{c1}$, $H_1 = 0.12$ T, and $H_W = 0.008$ T. (b) Magnetization hysteresis loop corresponding to the J_c values shown in (a). Hysteresis loop that would be obtained within the framework of critical state model, i.e., in the limit of $B_r \rightarrow 0$ is shown by the dotted line. Inset shows the functional form of B_r which is nonzero in the field range $H_{low} < H < H_p$.

The magnetization hysteresis loop corresponding to $J_c(H\uparrow)$ and $J_c(H\downarrow)$ is shown in Fig. 2(b). Note the asymmetry in the hysteresis, usually observed in experiments. For comparison, we also plot the magnetization hysteresis loop one would obtain within the framework of Bean's critical state model with $J_c = J_c^{st}$ (applicable in the limit $B_r \rightarrow 0$), which is symmetric in forward and reverse field cycles, as shown by the dotted line in Fig. 2(b). Details of the magnetization calculation are described in Ref. 20. The minor magnetization curves of the types I and II calculated in the slab geometry are shown in Figs. 3(a) and 3(b), respectively. They clearly mimic the behavior seen in experiments. We assumed $M_{eq}(H) = 0$, in calculating these magnetization curves. We note that the calculated curves in Figs. 2 and 3 are not quantitative fits to experimental data and only serve to illustrate qualitative features of the observed data.

In Fig. 3(c), we show $M_{eq}^*(H)$ determined from the calculated minor curves of types I and II following Eqs. (2) and (3), respectively. The test of the self-consistency of these procedures lies in reproducing the original form $(M_{eq} = 0)$ assumed in the calculation. $M_{eq}^*(H)$ obtained from these two procedures are not only inconsistent with each other but also do not conform to the original assumption that $M_{eq} = 0$.[21] The procedure of Eq. (2) indeed produces a peaklike structure in $M_{eq}^*(H)$, which has been shown earlier from an

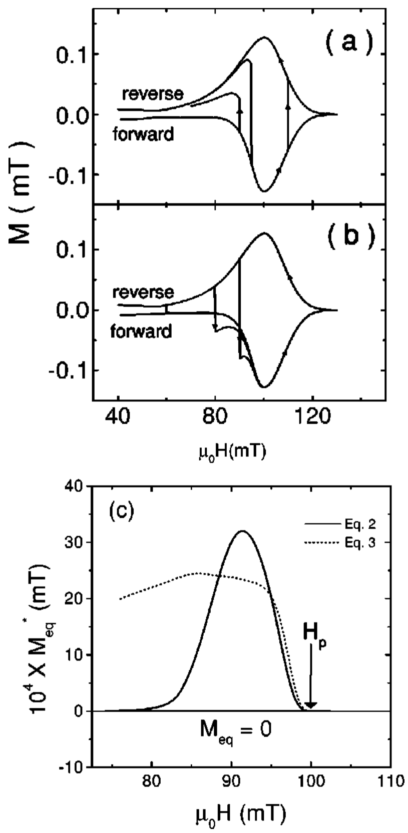

FIG. 3. Calculated minor curves of types I and II are shown in (a) and (b), respectively. In (c), we show the M_{eq}^{*} vs H obtained using Eqs. (2) and (3), respectively, along with the original form $M_{eq}=0$ assumed in the calculation of minor curves.

analysis of experimental data in $2H$-$NbSe_2$ following the same recipe.[19] On the other hand, the use of Eq. (3) proposed by Tenya *et al.*[18] yields no variation in M_{eq}^{*} vs H across the PE region. Figure 3(c) illustrates the unreliable and ambiguous nature of these recipes noted above and thus points to the need for a consistent approach in order to overcome their difficulties.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

In this section, we will show experimentally that repeated field cycling drives any metastable state into a stable state that is unique at a given field.[20] We study the minor hysteresis loops traced by repeated field cycling and infer from these measurements the critical current density J_c^{st} and the equilibrium magnetization M_{eq} of the stable state.

Direct current magnetization measurements have been carried out using a quantum design (QD) Inc. SQUID magnetometer (model MPMS5) in the peak-effect region of a $2H$-$NbSe_2$ single crystal ($T_c \approx 7.25$ K) with the field applied parallel to its c axis. The crystal is of approximate dimensions ($a \times b \times c$) $4 \times 5 \times 0.43$ mm. As stated earlier, the peak effect in J_c is manifested as the anomalous enhancement in the magnetization hysteresis (cf. Fig. 1). The magnetization hysteresis has been studied at different temperatures from 6.7 to 6.95 K. Magnetization hysteresis data at 6.95 K were measured using a 2-cm full-scan length, and the data at the other temperatures were obtained using the half-scan technique[12,22] to avoid artefacts arising due to field inhomogeneity experienced by the sample along the scan length. In the temperature range investigated, J_c at the peak field H_p decreases with decreasing temperature (see Table I).

TABLE I. Superconducting parameters in $2H$-$NbSe_2$.

T(K)	H_p(mT)	$\mu_0 \Delta M_{eq}(\mu T)/4\pi$	$J_c(H_p)$ (A/m^2)	$\Delta s(k_B)$
6.95	105	3.8 ± 0.4	54×10^4	13.6 ± 1.4
6.90	136	5.0 ± 0.4	36×10^4	13.8 ± 1.1
6.85	170	1.3 ± 0.4	26×10^4	2.9 ± 0.9
6.80	202	1.9 ± 0.4	17×10^4	3.5 ± 0.7

Figure 4(a) depicts a part of the hysteresis loop at 6.95 K, constituting M vs H curves in the increasing (forward) and decreasing (reverse) field cycles measured with a 30-s wait time at each field. We identify the onset field H_{pl}^{+} of the PE on the forward curve where M begins to decrease sharply. The field H_p marks the field at which magnetization hysteresis is maximum. In Fig. 4(a), we show points A, B, C, and D from where the minor hysteresis loops are initiated. A (C) and B (D) are at a field $H < H_{pl}^{+}$ ($H > H_{pl}^{+}$) on the forward and reverse curves, respectively. Minor hysteresis loops starting from both forward and reverse curves are recorded at different fields (spanning the peak region) by repeatedly cycling the field by a small amplitude ΔH. The interval ΔH is chosen such that it is above the threshold field required to reverse the direction of the shielding currents throughout the sample. From the critical state model, we understand that magnetization values must always remain confined within the forward and reverse magnetization curves, which constitute the so-called magnetization envelope. Further, the $M - H$ loop in each field cycle must retrace itself.

In Fig. 4(b), we show the minor hysteresis loops (MHLs) measured by repeatedly cycling the field, starting at point A ($H < H_{pl}^{+}$) on the forward curve. These MHLs in different field cycles retrace each other, indicating that the J_c does not change with field cycling. Therefore, we conclude that the vortex state is in a stable configuration. In contrast, the MHLs shown in Fig. 4(c) (continuous line with data points omitted) starting at B ($H < H_{pl}^{+}$) on the reverse curve, shrinks with each successive field cycle and finally collapses into the minor loop started from point A (open circles), which is replotted in Fig. 4(c). This suggests that the vortex configuration at point B is metastable with a $J_c > J_c^{st}$. Repeated field cycling causes the J_c to fall toward the stable value as reflected in the width of the MHLs collapsing with each successive field cycle. It is remarkable that the minor loops starting from both A and B merge into precisely the same loop within experimental accuracy. This clearly reaffirms the basic assumption of the model that there exists a unique stable state with critical current density J_c^{st}, independent of the initial vortex state from which it evolves.

We now focus on the behavior of MHLs that start from a field $H > H_{pl}^{+}$. As shown in Fig. 4(d), the behavior of the minor loops starting at point C is quite different from the

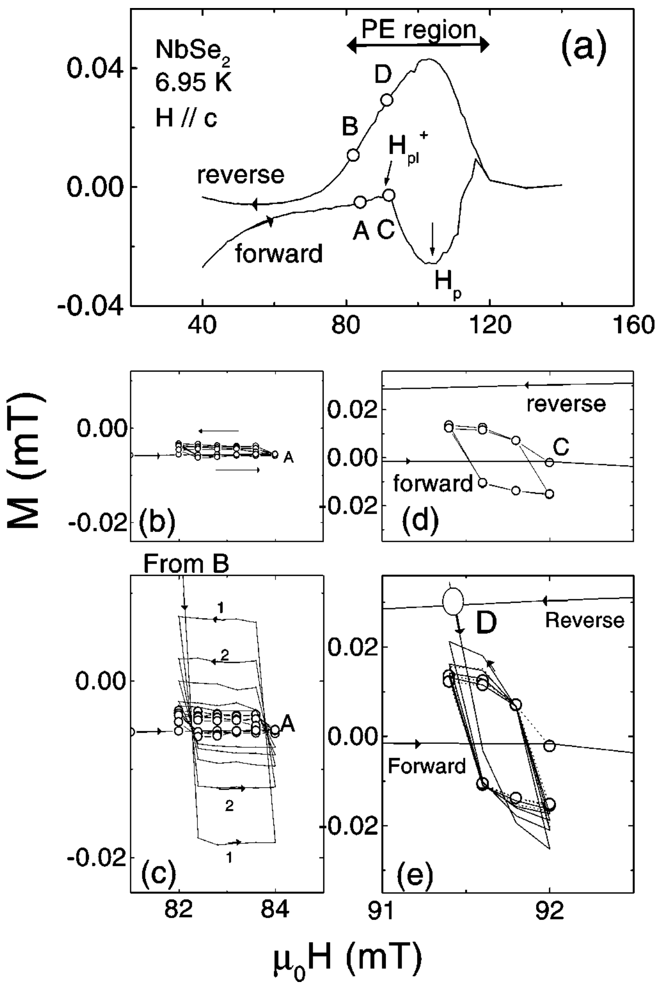

FIG. 4. (a) A part of the magnetization loop (forward and reverse curves) measured at 6.95 K on a $2H$-$NbSe_2$ single crystal. Also indicated are the characteristic fields H_{pl}^{+} and H_p. We indicate A and B ($H<H_{pl}^{+}$) and C and D ($H_{pl}^{+}<H<H_p$) starting from which the minor hysteresis loops are measured. (b) Minor hysteresis loops started from point A (open circles). In different field cycles, they are seen to retrace the same loop. (c) The MHL started from B (continuous line) shrinks with each successive field cycle. The increasing and decreasing field legs of the first and second cycles are numbered. After five field cycles, the hysteresis loop is seen to merge with the loop shown in (b), which is replotted (open circles). (d) Minor hysteresis loops started from point C (open circles). In the first field cycle itself, increasing field leg of the MHL separates from the forward curve and remains outside the magnetization envelope for the subsequent field cycles. (e) The minor loops starting from D (continuous line) are seen to collapse on to the loop shown in (d), which is replotted.

behavior started at point A. The increasing field leg of the MHL separates from the forward magnetization curve in the first field cycle itself and remains outside the magnetization envelope for subsequent field cycles. This clearly suggests that, for $H>H_{pl}^{+}$, the vortex configuration even on the forward magnetization curve is metastable. However, the behavior of the MHLs starting at point D on the reverse magnetization curve is very similar to the one that starts at point B, viz., the loop shrinks with each successive field cycle [continuous line in Fig. 4(e)]. The data in Fig. 4(d) are replotted in Fig. 4(e) (open circles connected by dotted line), which suggests that the MHLs starting from both C and D collapse into the same final loop. First, these data clearly suggest the metastable nature of the vortex configuration for fields above H_{pl}^{+} both on the forward and reverse magnetization curves. Further, the MHLs obtained on repeated field cycling is again independent of the initial vortex configuration. We note that the metastable state on the forward magnetization curve settles into the stable state much faster than that on the reverse curve. This might imply that the vortex configuration on the increasing field cycle is closer to the equilibrium configuration.

The data in Fig. 4 yield the following critical current inequalities in different field ranges. (i) For $H<H_{pl}^{+}$ vortex configuration is stable in the increasing field cycle, while at the same field value, it is highly metastable in the decreasing field cycle. This can be summarized by the inequality $J_c(H\uparrow)=J_c^{st}(H)<J_c(H\downarrow)$. (ii) For $H_{pl}^{+}<H<H_p$, vortex configurations in both increasing and decreasing field cycles are metastable, with the critical currents obeying the inequality $J_c(H\uparrow)<J_c^{st}(H)<J_c(H\downarrow)$. (iii) For $H>H_p$, $J_c(H\uparrow)=J_c(H\downarrow)=J_c^{st}(H)$. These observations are in accordance with the model[20] [cf. Fig. 2(a)]. We thus assert that Eq. (2), proposed by Roy and Chaddah[16] is applicable only for $H<H_{pl}^{+}$ and not for $H_{pl}^{+}<H<H_p$, as the vortex lattice on the forward curve is in a *superheated* vortex configuration that is more ordered (but metastable) than the stable configuration. Equation (3) as proposed by Tenya *et al.*[18] is not appropriate in any of the field ranges because the vortex state produced on the reverse curve is a *supercooled* vortex configuration,[14,23] which is more disordered than the stable state.

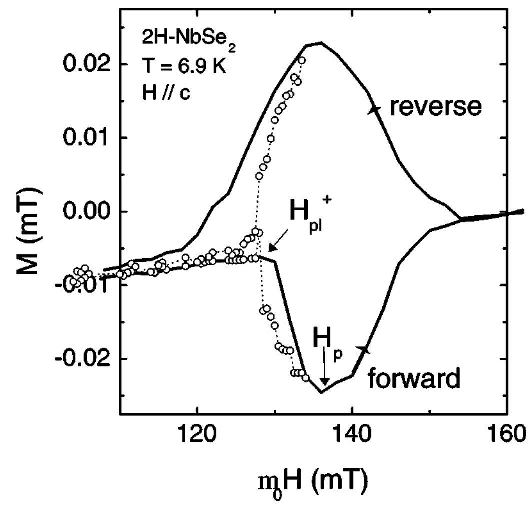

FIG. 5. Magnetization hysteresis loop of $2H$-$NbSe_2$ recorded using half-scan technique at 6.9 K (continuous line). The open circles are the saturated magnetization values obtained after repeated field cycling. H_{pl}^{+} and H_p are also marked. The locus of the saturated magnetization values is shown by a dotted line.

Figure 5 shows the M-H loop at 6.9 K constituting forward and reverse magnetization curves (dark line with data points omitted) indicating H_{pl}^{+} and H_p. Note the asymmetry

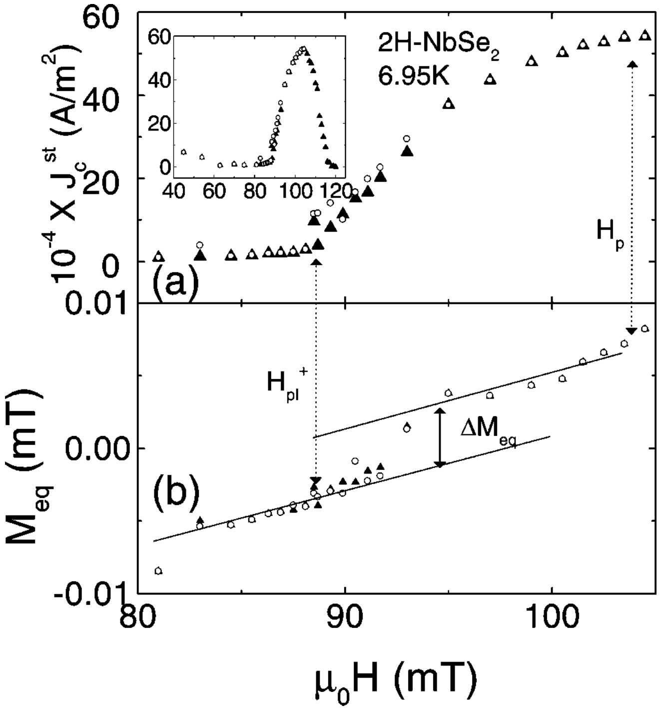

FIG. 6. (a) Stable critical current density J_c^{st} in the field range 80 mT$<\mu_0 H<$105 mT. In the inset we show the J_c^{st} vs H in the entire field range studied. Filled triangles and open circles correspond to the values obtained from the MHLs initiated from the forward and reverse magnetization curves, respectively. (b) Equilibrium magnetization M_{eq} as a function of field at 6.95 K. Note that the sharp change in M_{eq} coincides with the PE onset field H_{pl}^+.

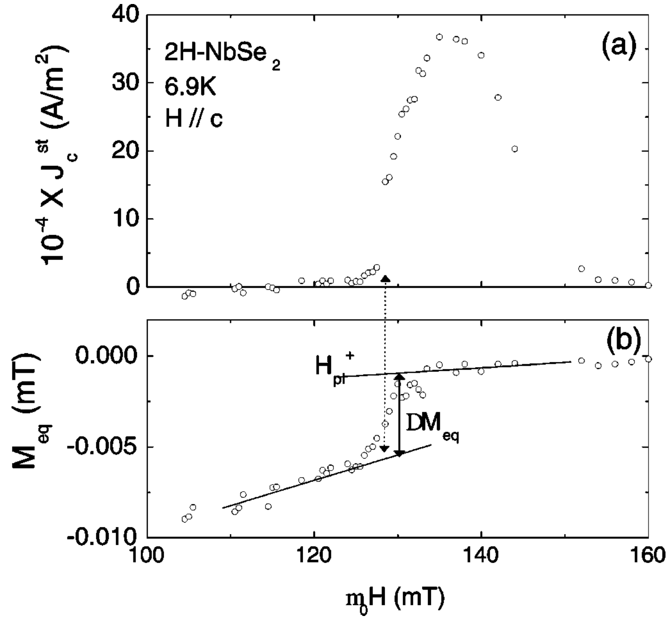

FIG. 7. (a) Critical current density J_c^{st} vs H and (b) M_{eq} vs H data obtained at 6.9 K. Note that the smeared jump in M_{eq} vs H, marked by the two-sided arrow, agrees precisely with a smeared jump in critical current density J_c^{st} in the peak regime. See the text for discussion.

(also seen at 6.95 K) in the forward and reverse magnetization curves that is the hallmark of the peak effect. We also measured the MHLs by repeatedly cycling the field starting at different points on the forward and reverse curves. The saturated MHLs are again found to be independent of the initial vortex state just as for 6.95 K. The locus of magnetization values on the increasing and decreasing field legs of the saturated MHLs measured at different fields is also plotted in Fig. 5 (open circles connected by dotted line). This is in excellent qualitative agreement with the behavior expected from the model in Ref. 20 [see Fig. 2(b)]. The locus of saturated magnetization values correspond to the stable or equilibrium vortex configuration at different fields.

Having established the existence of a history-independent stable state, we determine the critical current density J_c^{st} and equilibrium magnetization M_{eq} of the stable (or equilibrium) state at each field from the saturated MHL.[24] J_c^{st} and M_{eq} are given by

$$J_c^{st}(H)=[M_{st}(H\downarrow)-M_{st}(H\uparrow)]/2g\mu_0 R, \qquad (8a)$$

$$M_{eq}(H)=[M_{st}(H\uparrow)+M_{st}(H\downarrow)]/2, \qquad (8b)$$

where $M_{st}(H\uparrow)$ and $M_{st}(H\downarrow)$ are the magnetization values on the increasing and decreasing field legs of the saturated MHL. J_c^{st} vs H and M_{eq} vs H data at 6.95 K are plotted in Figs. 6(a) and 6(b), respectively. M_{eq} exhibits a sharp increase between H_{pl}^+ and H_p, signifying an increase in equilibrium flux density. This is reminiscent of the FLL melting transition observed in cuprate superconductors.[25,26] We argue that the change in M_{eq} indicates a first order transition in the FLL from an ordered solid to a pinned amorphous state,[19] presumably analogous to a Bragg Glass to Vortex Glass/pinned liquid phase transition.[27] The increase in M_{eq} coincides with the increase in J_c^{st} at the onset of the peak effect and spans the field range between H_{pl}^+ and H_p. In Figs. 7(a) and 7(b), we present the M_{eq} vs H and J_c^{st} vs H data, respectively, at 6.9 K. We note that the sharp change in M_{eq} correlates with a sharp increase in J_c^{st} between H_{pl}^+ and H_p. We also present the ΔM_{eq} values obtained at different temperatures in Table I. There seems to exist a correlation between the value of ΔM_{eq} and the $J_c(H_p)$.

Equation (8b) ignores any asymmetry in the induced currents that can arise from edge/surface effects.[28] For instance, above H_{pl}^+, disordered phase can be injected through the imperfections in the sample edges, resulting in a surface current distribution larger in the increasing field branch than that in the decreasing field branch. It can be easily seen that the asymmetry in the induced currents only suppresses the change in the M_{eq} above H_{pl}^+. In the absence of detailed knowledge of the edge effects, the ΔM_{eq} values presented in Table I serve as the lower limits for the ΔM_{eq} that are applicable in the bulk.

It is important to understand the nature of the vortex state in the transition region $H_{pl}^+<H<H_p$. One of the pictures is the collective pinning picture,[2] where the loss of long-range order is uniform throughout the sample. On the other hand, Paltiel *et al.*[28] proposed a picture where the disordered phase enters through surface imperfections and coexists at the surface with the ordered phase in the bulk. They argue that the

boundary between the disordered region and the ordered region moves into the sample as the temperature (or field) is increased toward T_p (or H_p). A further possibility is the coexistence of ordered and disordered phase with an intricate geometrical connectivity of these phases. Irrespective of the particular picture used, our experiments demonstrate a specific and unambiguous procedure, viz., subjecting the sample to a field cycling, to produce a unique stable state (in a macroscopic sense) across the peak-effect region.

We consider this stable state as a pinned equilibrium state and estimate equilibrium magnetization and the free-energy difference or entropy change when the vortex lattice changes from an ordered to an amorphous state. As per the Clausius-Clapeyron relation,[26,29] the entropy changes per vortex per interlayer distance d ($\simeq 4$ Å) in the $2H$-$NbSe_2$ system,[19]

$$\Delta s = -(\Delta M_{eq}/H_p)(dH_{pl}^{+}/dT)(\phi_0 d/k_B),$$

where $dH_{pl}^{+}/dT \simeq dH_p/dT \simeq -0.65$ T/K. The value of Δs estimated at different temperatures is tabulated in Table I. Incidentally, these values are comparable to the entropy change reported across the FLL melting transition in high-T_c cuprates.

An important question that can arise is whether the entropy change can be observed in thermal measurements such as specific heat versus temperature. We recall that the metastability in the vortex state is much greater in temperature scans in a fixed magnetic field.[14] Repeated cycling of field by a small amplitude may be necessary to produce the stable or equilibrium state before a thermal measurement is carried out at each temperature.

V. CONCLUSIONS

In this paper, we have presented a study of different metastable vortex configurations occurring in the peak-effect region of a weakly pinned superconductor $2H$-$NbSe_2$ through magnetization measurements. Each metastable vortex configuration is characterized by a critical current density J_c that is strongly dependent on the magnetic history. It is also shown that any metastable vortex configuration obtained under different field histories can be driven into a stable configuration by repeated field cycling. This stable configuration has a critical current density J_c^{st} uniquely determined by field and temperature as postulated in a recent model.[20] Field cycling appears to act as an effective temperature to drive a metastable state into the stable state even when thermal energy itself is inadequate to sample the phase space and access the stable state.

The method of recording minor hysteresis loops described here allows us to determine the pinning and equilibrium properties of the stable vortex state satisfactorily. Our equilibrium magnetization data clearly suggest that the transition of the vortex lattice from an ordered state to a disordered state is first order in nature. The smearing of the transition, i.e., the width of the transition region may be a manifestation of the spatially inhomogeneous pinning of the system. The J_c^{st} data suggest that the loss of quasi-long-range order in the vortex lattice also spans the same field window as the magnetization jump. In the collective pinning picture, this amounts to correlation volume of the vortex phase decreasing in this regime and the FLL becoming completely disordered above H_p or T_p. The precise coincidence of the J_c anomaly with the equilibrium magnetization anomaly further illustrates the self-consistency of the procedure developed here. It would be interesting to compare the nature of this disorder-driven transition in systems with different types of pinning, e.g., high density of point pins versus low density of extended pins to further understand the nature of this presumably disorder-induced phase transformation.

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[1] M. J. Higgins and S. Bhattacharya, Physica C **257**, 232 (1996), and references therein.

[2] A. I. Larkin and Y. N. Ovchinnikov, Zh. Éksp. Teor. Fiz **65**, 1704 (1973) [Sov. Phys. JETP **38**, 854 (1974)]; J. Low Temp. Phys. **34**, 409 (1979); A. I. Larkin, Zh. Éksp. Teor. Fiz. **58**, 1466 (1970) [Sov. Phys. JETP **31**, 784 (1970)].

[3] S. S. Banerjee, N. G. Patil, S. Saha, S. Ramakrishnan, A. K. Grover, S. Bhattacharya, G. Ravikumar, P. K. Mishra, T. V. Chandrasekhar Rao, V. C. Sahni, M. J. Higgins, E. Yamamoto, Y. Haga, M. Hedo, Y. Inada, and Y. Onuki, Phys. Rev. B **58**, 995 (1998).

[4] P. L. Gammel, U. Yaron, A. P. Ramirez, D. J. Bishop, A. M. Chang, R. Ruel, L. N. Pfeiffer, and E. Bucher, Phys. Rev. Lett. **80**, 833 (1998).

[5] T. V. Chandrasekhar Rao, V. C. Sahni, P. K. Mishra, G. Ravikumar, C. V. Tomy, G. Balakrishnan, D. Mck Paul, C. A. Scott, S. S. Banerjee, N. G. Patil, S. Saha, S. Ramakrishnan, A. K. Grover, and S. Bhattacharya, Physica C **299**, 267 (1998).

[6] C. P. Bean, Phys. Rev. Lett. **8**, 250 (1962); Rev. Mod. Phys. **36**, 31 (1964).

[7] W. A. Fietz and W. W. Webb, Phys. Rev. **178**, 657 (1969).

[8] P. Chaddah, S. B. Roy, and M. Chandran, Phys. Rev. B **59**, 8440 (1999).

[9] M. Steingart, A. G. Putz, and E. J. Kramer, J. Appl. Phys. **44**, 5580 (1973).

[10] R. Wordenweber, P. H. Kes, and C. C. Tsuei, Phys. Rev. B **33**, 3172 (1986).

[11] W. Henderson, E. Y. Andrei, M. J. Higgins, and S. Bhattacharya, Phys. Rev. Lett. **77**, 2077 (1996); **80**, 381 (1998).

[12] G. Ravikumar, V. C. Sahni, P. K. Mishra, T. V. Chandrasekhar Rao, S. S. Banerjee, A. K. Grover, S. Ramakrishnan, S. Bhattacharya, M. J. Higgins, E. Yamamoto, Y. Haga, M. Hedo, Y. Inada, and Y. Onuki, Phys. Rev. B **57**, R11 069 (1998).

[13] S. S. Banerjee, N. G. Patil, S. Ramakrishnan, A. K. Grover, S. Bhattacharya, G. Ravikumar, P. K. Mishra, T. V. Chandrasekhar Rao, V. C. Sahni, M. J. Higgins, C. V. Tomy, G. Balakrishnan,

and D. McK Paul, Phys. Rev. B **59**, 6043 (1999).

[14] G. Ravikumar, P. K. Mishra, V. C. Sahni, S. S. Banerjee, A. K. Grover, S. Ramakrishnan, P. L. Gammel, D. J. Bishop, E. Buscher, M. J. Higgins, and S. Bhattacharya, Phys. Rev. B **61**, 12 490 (2000).

[15] S. Sarkar, D. Pal, S. S. Banerjee, S. Ramakrishnan, A. K. Grover, C. V. Tomy, G. Ravikumar, P. K. Mishra, V. C. Sahni, G. Balakrishnan, D. McK Paul, and S. Bhattacharya, Phys. Rev. B **61**, 12 394 (2000).

[16] S. B. Roy and P. Chaddah, J. Phys.: Condens. Matter **9**, L625 (1997).

[17] S. Kokkaliaris, P. A. J. de Groot, S. N. Gordeev, A. A. Zhukov, R. Gagnon, and L. Taillefer, Phys. Rev. Lett. **82**, 5116 (1999).

[18] K. Tenya, S. Yasunami, T. Tayama, H. Amitsuka, T. Sakakibara, M. Hedo, Y. Inada, E. Yamamoto, Y. Haga, and Y. Onuki, J. Phys. Soc. Jpn. **68**, 224 (1999).

[19] G. Ravikumar, P. K. Mishra, V. C. Sahni, S. S. Banerjee, S. Ramakrishnan, A. K. Grover. P. L. Gammel, D. J. Bishop, E. Buscher, M. J. Higgins, and S. Bhattacharya, Physica C **322**, 145 (1999).

[20] G. Ravikumar, K. V. Bhagwat, V. C. Sahni, S. Ramakrishnan, A. K. Grover, and S. Bhattacharya, Phys. Rev. B **61**, R6479 (2000).

[21] For $H > H_p$, Eq. (1b) is more appropriate for obtaining M_{eq} because J_c is independent of magnetic history. This would give $M_{eq} = 0$.

[22] G. Ravikumar, T. V. Chandrasekhar Rao, P. K. Mishra, V. C. Sahni, S. S. Banerjee, A. K. Grover, S. Ramakrishnan, S. Bhattacharya, M. J. Higgins, E. Yamamoto, Y. Haga, M. Hedo, Y. Inada, and Y. Onuki, Physica C **298**, 122 (1998); G. Ravikumar, T. V. Chandrasekhar Rao, P. K. Mishra, V. C. Sahni, Subir Saha, S. S. Banerjee, N. G. Patil, A. K. Grover, S. Ramakrishnan, S. Bhattacharya, E. Yamamoto, Y. Haga, M. Hedo, Y. Inada, and Y. Onuki, Physica C **276**, 9 (1997).

[23] S. B. Roy and P. Chaddah, Pramana, J. Phys. **53**, 659 (1999).

[24] The critical current density $J_c = \delta M / \Omega$, where δM is the magnetization hysteresis expressed in units of A m and $\Omega = 10^{-3} a(1 - a/3b)/4$. See M. Jirsa *et al.*, *Proceedings of the 7th International Workshop on Critical Currents in Superconductors, Alpbach, Austria*, edited by H. W. Weber (World Scientific, Singapore, 1994), p. 221.

[25] H. Pastoriza, M. F. Goffman, A. Arribere, and F. de la Cruz, Phys. Rev. Lett. **72**, 2951 (1994).

[26] E. Zeldov, D. Majer, M. Konczykowski, V. B. Geshkenbein, and V. M. Vinokur, Nature (London) **375**, 373 (1995).

[27] T. Giamarchi and P. Le Doussal, Phys. Rev. Lett. **72**, 1530 (1994); Phys. Rev. B **52**, 1242 (1995); **55**, 6577 (1997).

[28] Y. Paltiel, E. Zeldov, Y. N. Myasoedov, H. Shtrikman, S. Bhattacharya, M. J. Higgins, Z. L. Xiao, E. Y. Andrei, P. L. Gammel, and D. J. Bishop, Nature (London) **403**, 398 (2000).

[29] U. Welp, J. A. Fendrich, W. K. Kwok, G. W. Crabtree, and B. W. Veal, Phys. Rev. Lett. **76**, 4809 (1996).