Paramagnetic magnetization signals and curious metastable behaviour in field-cooled magnetization of a single crystal of superconductor 2H-$NbSe_2$

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Abstract

We present here some newer characteristics pertaining to paramagnetic Meissner effect like response in a single crystal of the low T_c superconducting compound 2H-$NbSe_2$ via a detailed study of effects of perturbation on the field-cooled magnetization response. In the temperature range, where an anomalous paramagnetic magnetization occurs, the field-cooled magnetization response is found to be highly metastable: it displays a curious tendency to switch randomly from a given paramagnetic value to a diamagnetic or to a different paramagnetic value, when the system is perturbed by an impulse of an externally applied ac field. The new facets revealed in a single crystal of 2H-$NbSe_2$ surprisingly bear a marked resemblance with the characteristics of magnetization behaviour anticipated for the giant vortex states with multiple flux quanta ($L\Phi_0$, $\Phi_0 = hc/2e$, $L > 1$) predicted to occur in mesoscopic-sized superconducting specimen and possible transitions amongst such states.

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I. INTRODUCTION

The observation of an anomalous paramagnetic magnetization signal (viz., paramagnetic Meissner effect (PME)), instead of the usual diamagnetic behaviour, on field - cooling a superconducting specimen, continues to attract [1, 2] attention ever since its discovery [3] in ceramic sample(s) of a high T_c cuprate. In addition to high T_c cuprates [3–12], the PME-like attribute is known to occur in a wide variety of other superconductors [13–26]. Numerous explanations (see Ref. [27] for a review), e.g., the d - wave superconductivity [9], orbital glass [27, 28], presence of π contacts leading to spontaneous currents [4, 8, 29], Josephson junctions [30], etc., have been advanced as the origin of PME like signal(s) in high T_c superconductors. A model proposed by Koshelev and Larkin [31] envisaged the possibility of trapping of magnetic flux in the interior of a superconducting sample due to inhomogeneous cooling. In such a circumstance, the PME can occur due to unbalancing between two oppositely directed currents, viz., (i) a (paramagnetic) current set up in the interior that attempts to screen the effect of trapped flux and, (ii) a usual (diamagnetic) current flowing on the surface that shields the external field [31]. However, within the Ginzburg - Landau framework, the theoretical works by Moshchalkov *et al.* [32] and Zharkov [33], for model cases of mesoscopic cylindrical shaped superconductors, lead to the proposition that the multi - quanta vortex matter, i.e., vortices with multiple flux quanta, $L\Phi_0$, $\Phi_0 = hc/2e$, $L > 1$, nucleating below a third critical field (H_{c3}), at the onset of surface superconductivity [34] can also give rise to the PME.

Recently, a controlled switching of the PME into the usual (diamagnetic) Meissner effect has been vividly demonstrated by Xing *et al.* [26] in a superconducting ferromagnet Pb-Co nanocomposite, wherein the source of PME is argued to be related to some different mechanism other than those stated above. This manipulation of PME is possible by the change in orientation of magnetic moments of Co nanoparticles with respect to the external magnetic field [26]. Such a tuning of the anomalous PME and the non-anomalous (diamagnetic) Meissner effect has rarely been reported in the literature. As another case of manipulating a PME like response, we have explored some novel facets of the PME in a single crystal of 2H-$NbSe_2$ [35], an example of a conventional low T_c superconducting system often studied to explore novel notions in vortex matter. The results being reported ahead in 2H-$NbSe_2$ present an advancement over studies earlier reported by some of us in $Ca_3Rh_4Sn_{13}$ [24] and Nb [17]. The curious behaviour pertains to an unusual paramagnetic magnetization (*a la* PME) and its manipulation just below T_c in the (isofield) temperature-dependent dc magnetization ($M(T)$) data. We find that in a short window of temperature, where the PME occurs below T_c in 2H-$NbSe_2$, an external perturbation can randomly change the magnetization from a given paramagnetic value to a larger or lesser paramagnetic value or from a given paramagnetic state to a diamagnetic one. Across the same temperature interval, the system can also transit from a given diamagnetic state into a paramagnetic one after being perturbed by an ac field impulse. The vortex matter in the field-temperature domain of the PME in 2H-$NbSe_2$ is thus found to be highly metastable. The results in a bulk 2H-$NbSe_2$ specimen unexpectedly appear to echo the consequences of nucleation of several

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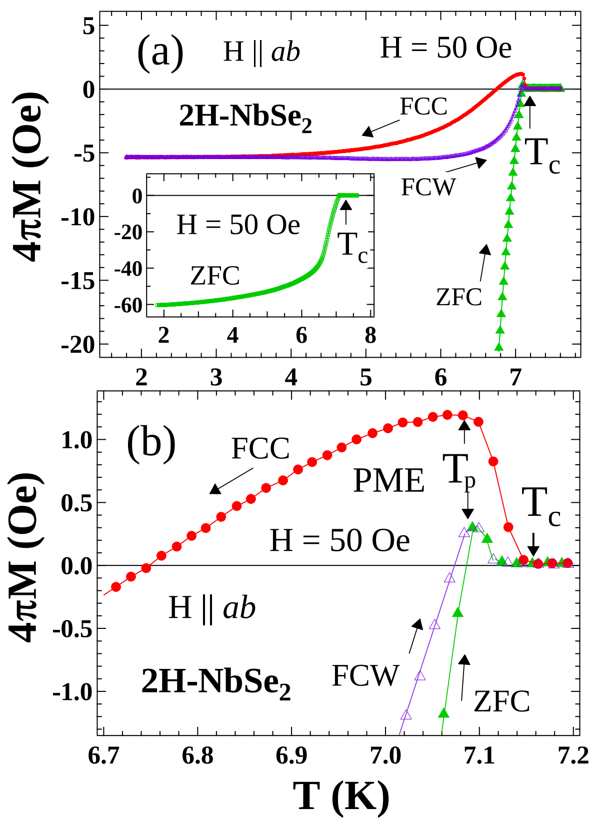

Figure 1: (Color online) (a) Temperature variation of M_{ZFC}, M_{FCC} and M_{FCW} in a field of H $(\parallel ab) = 50\,$Oe. The inset in panel (a) illustrates $M_{ZFC}\,(T)$ plot in the range, $1.8\,$K $< T < T_c$. Panel (b) elucidates $M(T)$ plots on the expanded scale in the vicinity of T_c. Anomalous paramagnetic signals are evident in all three modes. The paramagnetic magnetization reaches its extreme limit at T_p during the field-cooled cool-down run, $M_{FCC}\,(T)$.

metastable multiple flux quanta states ($L > 1$) proposed in the context of mesoscopic samples [32, 33, 36–40].

II. EXPERIMENTAL DETAILS

The single crystal of hexagonal 2H-NbSe$_2$ chosen for the present study is platelet-shaped with a planar area of $4.12\,\text{mm}^2$, thickness of $0.17\,$mm and mass of $4.64\,$mg. The superconducting transition temperature (T_c) for this crystal is measured to be about $7.15\,$K. The crystallographic ab plane of the hexagonal 2H-NbSe$_2$ coincides with the plane of the platelet, while the direction normal to it corresponds to the crystallographic c - axis of 2H-NbSe$_2$. The magnetization measurements were performed using a Superconducting Quantum Interference Device - Vibrating Sample Magnetometer (SQUID - VSM, Quantum - Design, Inc., USA) in such a way that the magnetic field was directed nominally parallel to the plane of the platelet (i.e., $H \,||\, ab$), with less than 5 degree error in the angle of alignment. An advantage of investigating the sample along this orientation is that the demagnetization factor is small ($\sim 10^{-1}$) and the associated geometrical/boundary effects are expected to be minimal. We estimated, using the flux gate option of the SQUID - VSM, the field-inhomogeneity of the superconducting magnet over a scan length of $8\,$mm. It is found to be only of the order $10^{-2}\,$Oe. During the magnetization measurements, the amplitude of vibration of the sample was kept small ($\approx 0.5\,$mm) in most of the runs (unless specifically stated otherwise) so that the possible artefact [41] due to field inhomogeneity along the scan length of the superconducting sample in SQUID - VSM is so minuscule that it is of no significance. The effect of changing the amplitude of sample vibration on the magnetization value (at a given H, T value) in the superconducting state of a standard Indium sample has also been investigated. We registered no difference in the magnetization response of Indium specimen in its superconducting state, when the amplitude of vibration in VSM measurements was changed from 0.5 to $8\,$mm. Hence, under the normal circumstances, it can be stated that the change in amplitude of sample vibration (upto a maximum of $8\,$mm in the present VSM instrument) is not expected to alter the dc magnetization value of a superconducting sample at a given H and T. In the case of 2H-NbSe$_2$ crystal, we have observed a wide variation in (isofield) magnetization response, ranging from paramagnetic to diamagnetic values at different amplitudes (see Fig. 7), which is argued to be a consequence of metastability effects prevailing in this system, a key focus of the present report.

III. RESULTS

A. Paramagnetic signal on field - cooling

Figure 1 displays the isofield magnetization responses obtained in the zero-field cooled ($M_{ZFC}\,(T)$), field-cooled cool-down ($M_{FCC}\,(T)$) and field-cooled warm-up ($M_{FCW}\,(T)$) modes in 2H-NbSe$_2$ in a field of $50\,$Oe. For $M_{ZFC}\,(T)$ run, the crystal was initially cooled down to $1.8\,$K in nominal zero field. A field, $H = 50\,$Oe, was then applied (such that $H \parallel ab$) and the magnetization obtained while warming to higher temperatures, yielding $M_{ZFC}\,(T)$. Without changing the magnetic field, the $M(T)$ data were again obtained while cooling the sample back to the lowest temperature ($M_{FCC}\,(T)$). Thereafter, the magnetization values were measured again while warming it to yield $M_{FCW}\,(T)$. The $M_{ZFC}\,(T)$ curve shows the usual fall in diamagnetic response (cf. inset panel of Fig. 1(a)) of a superconducting specimen as the temperature is swept up.

An expanded portion of $M(T)$ plots of Fig. 1(a), in the proximity of T_c is displayed in Fig. 1(b). Unusual paramagnetic responses below T_c can be observed in all the three modes. It is pertinent to note that the paramagnetic signals in $M_{ZFC}\,(T)$ and $M_{FCW}\,(T)$ runs are restricted to a very narrow temperature interval ($\sim 50\,$mK) a little below T_c. During $M_{FCC}\,(T)$ run, the paramagnetic response nucleating at T_c enhances as the temperature is lowered towards a temperature marked as T_p. Thereafter, the $M_{FCC}\,(T)$ values start to decrease, and cross over to the diamagnetic values at a temperature of about 6.75 K. The diamagnetic $M_{FCC}\,(T)$ signal reaches a saturated value below 4 K (cf. main panel of Fig. 1(a)). During warm-up run, saturated $M_{FCW}\,(T)$ values remain sustained upto about 6 K. Above this temperature, the diamagnetic $M_{FCW}\,(T)$ values decrease, and they crossover to the paramagnetic values to merge into the $M_{ZFC}\,(T)$ curve at about 7.08 K, which is nearly the same temperature (marked as T_p) below which paramagnetic signal starts to decrease during the corresponding $M_{FCC}\,(T)$ run. It can be noted in Fig. 1(b), that the paramagnetic response in $M_{FCC}\,(T)$ run is much larger than that in $M_{ZFC}\,(T)$ run or $M_{FCW}\,(T)$ run. This observation implies that the positive magnetization signal below T_c is multi-valued (and thus metastable) and depends significantly on the thermomagnetic history of the sample.

Figure 2 illustrates the $M_{FCC}\,(T)$ curves for $H\,(\parallel ab) =$ 100 Oe, 500 Oe and 1 kOe, which intersect unexpectedly at a temperature, marked as T_{cross} ($\approx 6.84\,$K), where the magnetization incidentally also crosses the $M = 0$ axis. Below T_{cross}, the magnetic behaviour displayed in Fig. 2 is such that the diamagnetic response decreases as the field increases ($(\triangle M/\triangle H) > 0$), which is typical of a vortex state of a type - II superconductor in the field interval, $H_{c1} < H < H_{c2}$. On the other hand, the magnetization response curves above T_{cross} imply that the paramagnetism decreases as the applied field increases [32].

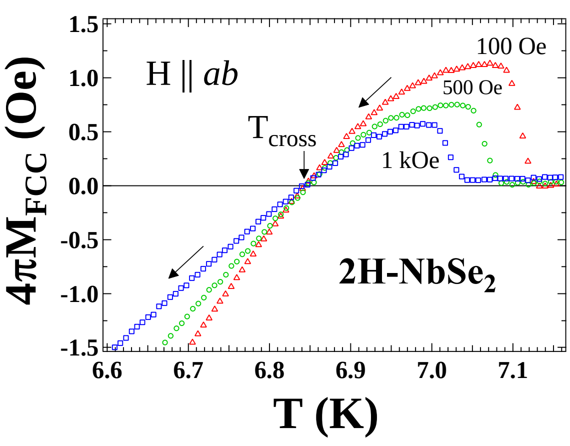

Figure 2: (Color online) Portions of the $M_{FCC}\,(T)$ curves at $H = 100\,$Oe, 500 Oe and 1 kOe, intersecting unusually at a characteristic temperature T_{cross}.

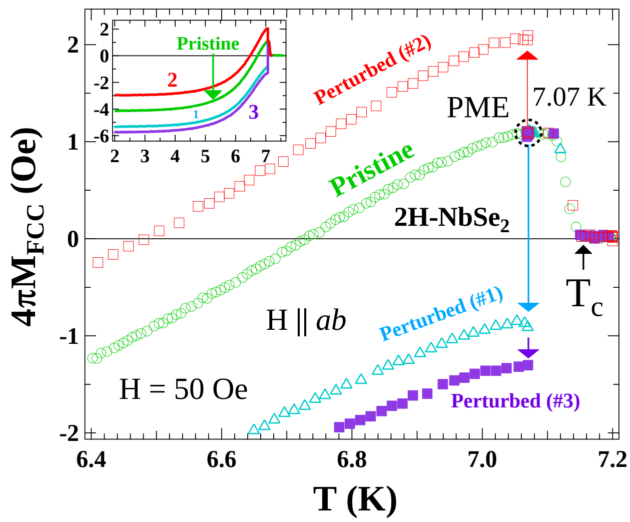

Figure 3: (Color online) $M_{FCC}\,(T)$ data recorded at $H = 50\,$Oe in several runs. In each of these runs, an impulse of an ac field (peak amplitude = 10 Oe, frequency = 211 Hz) was applied at $T = 7.07\,$K (encircled), after which the field-cooled magnetization change randomly to either paramagnetic or diamagnetic values, and $M_{FCC}\,(T)$ traverses different paths as displayed for three different runs, viz., perturbed (#1), perturbed (#2) and perturbed (#3). The pristine $M_{FCC}\,(T)$ (open circles) was recorded without perturbing the system throughout the cooling. The inset displays $M_{FCC}\,(T)$ for pristine and perturbed (#1, #2, and #3) cases in the full range, $2\,\mathrm{K} < T < 7.5\,\mathrm{K}$.

B. Metastability and non - uniqueness in magnetization response

The multi - valuedness in $M(T)$ observed in Fig. 1(b) motivated further exploration of magnetization response in 2H-NbSe$_2$. We illustrate in the main panel of Fig. 3, portions of $M_{FCC}\,(T)$ curves in the vicinity of T_c recorded at $H = 50\,$Oe during several runs where a perturbation had been induced at an intermediate stage. First, we recorded the magnetization data (see open circles in Fig. 3) while field-cooling the sample to 2 K, similar to the way $M_{FCC}\,(T)$ data of Fig. 1 were obtained. This curve will henceforth be called as "pristine" in the discussion ahead. We then recorded the $M_{FCC}\,(T)$ data (without changing the external field) while cooling the sample down to (near) peak temperature (T_p) of the PME, i.e., $T \approx 7.07\,$K, which is encircled in the main panel of Fig. 3. At this temperature, the $M_{FCC}\,(T)$ measurements were paused and the system was momentarily subjected to a perturbation in the form of an ac field impulse of peak amplitude 10 Oe (frequency of ac field = 211 Hz) imposed for a duration of 6 seconds. It was checked that the results being reported in Fig. 3 do not depend on the duration of the impulse. The $M_{FCC}\,(T)$ data recorded after the impulse treatment showed an abrupt switching from paramagnetic to diamagnetic value, which is indicated by an arrow pointing towards open triangles in main panel of Fig. 3. The $M_{FCC}(T)$ curve (open triangles labeled as "perturbed (#1)") thereafter traverses a path which is akin to the one that seems to display the usual (diamagnetic) Meissner effect in contrast to the anomalous PME observed for the pristine curve (open circles). In the next run, we again cooled the sample down to the same temperature, T_p ($\approx$ 7.07 K), and once again applied the same impulse. Surprisingly, the $M_{FCC}(T)$ data now displayed (see perturbed (#2) in Fig. 3) an enhancement in the paramagnetic signal (shown by upward arrow pointing to open squares in Fig. 3). This is in sharp contrast to the impact of an impulse in the "perturbed (#1)" case, which involved switching of the magnetization from paramagnetic to a diamagnetic value. On further lowering of temperature, the $M_{FCC}(T)$ curve for perturbed (#2) can be seen to crossover to diamagnetic values at a temperature ($\approx$ 6.48 K), which is lower than the corresponding crossover temperature of the pristine $M_{FCC}(T)$ curve. In the identical imposition of an ac impulse in the third run, the $M_{FCC}(T)$ data (perturbed (#3)) can be switched from paramagnetic to a diamagnetic value which is even larger than that during perturbed (#1). On progressively lowering the temperature below T_p, the $M_{FCC}(T)$ curve for "perturbed (#3)" (closed squares) is seen to stay more diamagnetic (at a given T) than that observed in the "perturbed (#1)", as shown in Fig. 3.

In the inset panel of Fig. 3, we show the $M_{FCC}(T)$ curves for the pristine and perturbed states (#1, #2 and #3) in the entire temperature range of investigation, i.e., 2 K$< T <$ 7.5 K. It is curious to note that the saturated $4\pi M$ value at 2 K in a given perturbed run roughly amounts to the summation of the saturated $4\pi M$ for the pristine curve and the change in magnetization, $\triangle(4\pi M)$ induced at T_p due to an impulse treatment.

Subsequent efforts of applying the impulse not only at T_p but also in its vicinity, viz., across 6.75 K$< T < T_c$ (all data not shown here), lead to the inference that the $M_{FCC}(T)$ value, after perturbation in the PME region, can unpredictably change very widely from paramagnetic to diamagnetic or from a given paramagnetic to a less/more paramagnetic value. We also applied impulse with different amplitudes and at different frequencies of the ac field, however, the results turned out to be same. In all, we can surmise that the vortex matter in the temperature regime of PME in 2H-NbSe$_2$ is highly metastable and the magnetization associated with such metastable states can show a rich diversity, as is evidenced in Fig. 3. When subjected to a perturbation, the system can transit between these metastable states, as is reflected from abrupt change(s) in magnetization.

C. Path dependent $M(T)$ response

We now demonstrate in Fig. 4, the impact of an ac field impulse applied at various temperatures, during a given

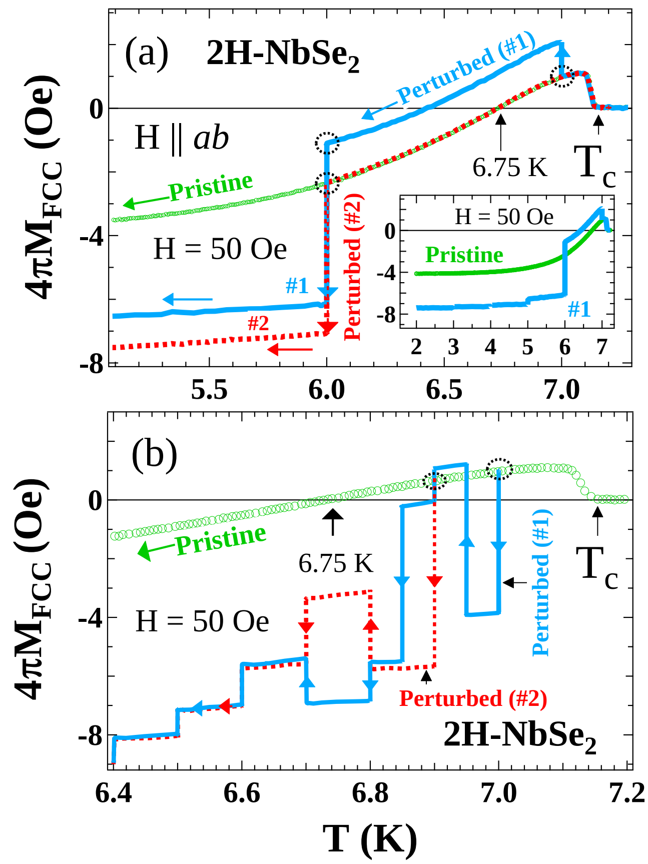

Figure 4: (Color online) $M_{FCC}(T)$ recorded at H = 50 Oe in various runs perturbed at different temperatures. (a) Perturbed (#1) shows an enhanced paramagnetism at 7 K and an enhanced diamagnetism at 6 K when compared with the pristine $M_{FCC}(T)$. In case of perturbed (#2), an impulse at 6 K displays equal change in magnetization as is seen in case of perturbed (#1). Inset shows pristine and perturbed (#1) in the full temperature range. (b) $M_{FCC}(T)$ traced while an impulse is applied at short temperature interval in two runs, perturbed (#1) and perturbed (#2), leading to a characteristic path dependence above 6.75 K, whereas below it, both follows the same path.

$M_{FCC}(T)$ run at H = 50 Oe. The pristine $M_{FCC}(T)$ shown in Fig. 4 is the same as presented in Fig. 3. When an impulse was applied at T = 7 K (encircled in Fig. 4(a)), an enhanced paramagnetic signal was seen as depicted by the $M_{FCC}(T)$ curve named "perturbed (#1)". This curve crosses over to the diamagnetic region smoothly at $T \sim$ 6.4 K, and thereafter remains less diamagnetic (than the pristine $M_{FCC}(T)$) until about T = 6 K, echoing the "perturbed (#2)" case of Fig. 3. At T = 6 K, an impulse was imposed again (see the upper encircling done corresponding to 6 K in Fig. 4(a)) and the M_{FCC} curve thereafter (i.e., perturbed (#1)) becomes more diamagnetic than the pristine one. The field-cooled curve (perturbed (#1)) follows a path shown by a continuous line below 6 K in Fig. 4(a),which is different from its pristine counterpart. The saturated $M_{FCC}(T)$ value at 2 K corresponding to perturbed (#1) curve is found to be nearly twice the saturated $M_{FCC}(T)$ in the pristine case, as is evident in the inset panel of Fig. 4(a). In another run, we applied an impulse once again at 6 K, but on the pristine field-cooled (FC) state (see another encircling done at 6 K in Fig. 4(a)). Such an impulse treatment resulted in the diamagnetic value of pristine $M_{FCC}(T)$ to switch over to a still larger diamagnetic value. The $M_{FCC}(T)$ curve thereafter, i.e., perturbed (#2) shown by the dashed line in Fig. 4(a), can be seen to be even more diamagnetic than the other two curves. We also found that after several iterations, that the change in magnetization induced due to impulse treatments below about 6.7 K is only of one kind, i.e., from a given diamagnetic value to another diamagnetic value, unlike the behaviour noticed above 6.75 K (i.e., in the PME region), where one could see switching from para/diamagnetic to more or less para/diamagnetic value (see Fig. 3).

In Fig. 4(b), we show the $M_{FCC}(T)$ curves at $H = 50$ Oe, when an ac field impulse is consecutively imposed at short temperature interval(s) in a given FC run. We first applied an impulse at $T = 7$ K (encircled in Fig. 4(b)), and observed a switching of paramagnetic M_{FCC} of the pristine state into a diamagnetic value (perturbed (#1)) as shown by continuous line. This $M_{FCC}(T)$ curve (perturbed (#1)) traces the record while cooling the sample further down from 7 K to 6.95 K, where the application of another impulse switches the diamagnetic value into a paramagnetic value, which is even more paramagnetic than the pristine one. As the cooling was progressed to $T = 6.9$ K, an impulse treatment there lead to the change from paramagnetic M_{FCC} value to near zero magnetization value. This process of applying an ac impulse at short intervals was continued, and the $M_{FCC}(T)$ curve (#1) displayed in Fig. 4(b) was traced. In another field-cooling run, we applied the impulse for the first time at $T = 6.9$ K, which resulted in switching from paramagnetic to a diamagnetic value as shown by the dashed curve (perturbed (#2)) in Fig. 4(b). This curve was recorded while further cooling, it also comprised impulse treatment(s) at several intermediate temperatures (see steps in magnetization where the impulse was applied) during the cool-down process.

An examination of $M_{FCC}(T)$ data recorded in the two runs, "perturbed (#1)" and "perturbed (#2)", in Fig. 4(b) shows a pronounced path-dependence in $M_{FCC}(T)$ at temperatures above about 6.7 K. This suggests that the impact of an impulse above 6.7 K is completely unpredictable: the system can display any magnetization value ranging from paramagnetic to diamagnetic. The impulse seems to be acting as a drive which brings out transition from a given (paramagnetic/diamagnetic) state of the system into another and hence leads to a non-uniqueness in $M_{FCC}(T)$. On the other hand, the two $M_{FCC}(T)$ runs ("perturbed (#1)" and "perturbed (#2)") tend to overlap when an impulse was applied below about 6.7 K. Below this temperature, the two curves

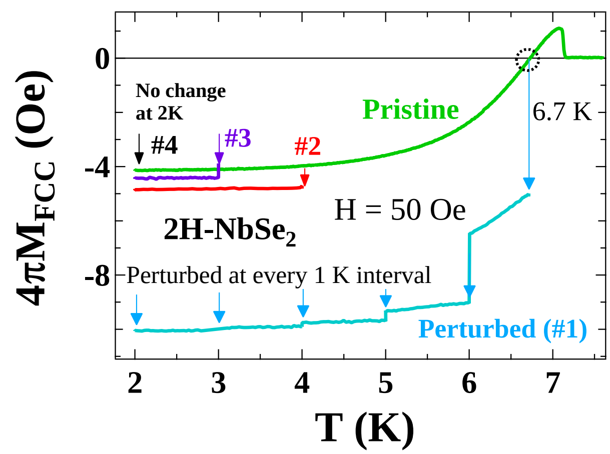

Figure 5: (Color online) $M_{FCC}(T)$ in pristine and perturbed cases at $H = 50$ Oe. In perturbed (#1), an impulse was first imposed at $T = 6.7$ K (encircled), followed by another at 6 K, and at every 1 K difference below 6 K. Saturated $M_{FCC}(T)$ value of pristine and perturbed (#1) are much different. In contrast, the impulse applied at 4 K (perturbed (#2)) and at 3K (perturbed (#3)) brings out minimal changes. No change is registered when the impulse is applied at 2 K.

respond identically to any further impulse treatment and the magnetization values remain diamagnetic.

To explore the highest saturated value of diamagnetic response, we chose to successively apply an ac field impulse below 6.7 K at every 1 K interval beginning from the pristine field-cooled (FC) state created at $T \approx 6.7$ K as shown in Fig. 5. The arrows in Fig. 5 indicate the temperatures at which the impulse was applied. The first impulse applied at 6.7 K changes the magnetization from a diamagnetic value (encircled) of pristine $M_{FCC}(T)$ to a larger diamagnetic value (perturbed (#1)). After lowering the temperature to 6 K, another impulse drives the magnetization into a still more diamagnetic state and so on. The saturated $4\pi M$ value for the perturbed (#1) $M_{FCC}(T)$ curve in Fig. 5 at 2 K happens to be about -10 Oe, which is more than twice the saturated $4\pi M$ value (≈ -4 Oe) of the pristine curve.

The commencement of application of an impulse at temperatures lower than 4 K resulted in a small change in the magnetization. For example, the changes in magnetization after the impulse treatment at $T = 4$ K and 3 K in pristine $M_{FCC}(T)$ runs were comparatively smaller in the perturbed states "#2" and "#3", respectively. Eventually, at $T = 2$ K (perturbed (#4)), no change in pristine M_{FCC} was witnessed after the application of an impulse at this temperature (cf. Fig. 5).

The results in Figs. 4 and 5 focus on $M_{FCC}(T)$ data, while subjecting the sample to perturbations at intermediate temperatures during cool-down cycle. In Fig. 6, we show another set of magnetization data, wherein we present the $M_{FCW}(T)$ responses recorded while sweeping the temperature up after an impulse treatment to (pristine) FC states created at various temperatures. In the first run, after getting an enhanced paramagnetism for impulse applied at $T = 7\,\mathrm{K}$ (encircled in main panel of Fig. 6), we recorded the magnetization (perturbed (#1)) while warming the sample towards the normal state. The paramagnetic values for "perturbed (#1)" fall smoothly as the temperature approaches T_c, however, this curve does not merge with the pristine $M_{FCC}\,(T)$ data. Magnetization data in the "perturbed (#2)" (open triangles) and "perturbed (#3)" (stars) states showed a switching towards diamagnetic region after the impulses were applied at 7 K and 6.71 K, respectively. While warming-up, the M_{FCW} curve for "perturbed (#3)" merges with that of "perturbed (#2)" at $T = 7\,\mathrm{K}$, and the two curves together follow nearly the same path thereafter. It is a mere coincidence that the first magnetization data after the impulse treatment at 7 K in the case of perturbed (#2) falls on the warm-up curve of perturbed (#3). Taking cue from the observations made in Fig. 3, the magnetization after the impulse treatment at 7 K could have achieved any value ranging from paramagnetic to diamagnetic. Some examples of path-dependent $M_{FCW}\,(T)$ responses in the close proximity of T_c are displayed in an inset panel of Fig. 6. In the fourth run, we applied the impulse at $T = 7.10\,\mathrm{K}$, and recorded the $M_{FCW}\,(T)$ thereafter while warming up the sample (see M_{FCW} for "perturbed (#4)" in the inset of Fig. 6). This curve can be seen to follow a path different from the pristine cooldown one, as is apparent from the inset panel. When we once again applied the impulse at $T = 7.10\,\mathrm{K}$ i.e., the 5^{th} attempt, the $M_{FCW}(T)$ curve (perturbed (#5)) nearly traced the path of the pristine M_{FCC} curve. Thus, during the warm-up runs also, one can observe a variety and path dependence in $M(T)$ response after creating different perturbed states (see "#1" to "#5" in Fig. 6).

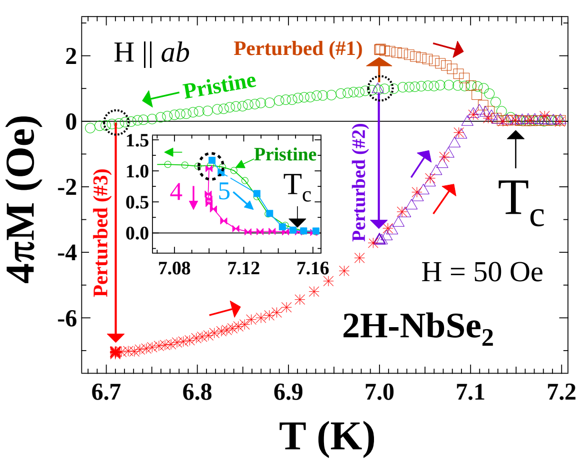

Figure 6: (Color online) $M_{FCW}\,(T)$ curves at $H = 50\,\mathrm{Oe}$ traced in five different warm-up runs after perturbation by an ac field impulse (perturbed (#1) to (#5)). The pristine curve (open circles) which was recorded during field-cooling in same field is also shown. Magnetization data in the perturbed conditions (#1 to #5) were recorded while warming the sample to higher T, after perturbing the pristine FC state created at $T = 7\,\mathrm{K}$ (perturbed (#1) and perturbed (#2)), $T = 6.71\,\mathrm{K}$ (perturbed (#3)) and $T = 7.1\,\mathrm{K}$ (inset shows perturbed (#4) and (#5)). In all the five cases, the $M_{FCW}\,(T)$ curves show a variety of paths that can be traversed while warming after the perturbation.

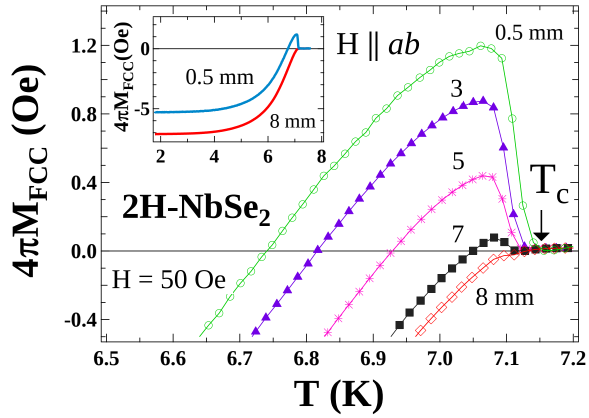

Figure 7: (Color online) Portions of the $M_{FCC}\,(T)$ curves recorded in $H = 50\,\mathrm{Oe}$ at various amplitudes of vibration in a VSM, as indicated. The saturated values of M_{FCC} at $T \sim 1.8\,\mathrm{K}$ for two of the curves, namely those recorded for 0.5 mm and 8 mm, can be seen to be different in the inset panel.

D. Diversity in M_{FCC} across the region of PME

We have so far restricted our discussion to the effect of a perturbation in the form of an ac field impulse on the FC magnetization response (Figs. 3 to 6). It is also tempting to ask whether a multiplicity in $M(T)$ similar to that observed in Figs. 3 to 6 can also be explored by any other form of perturbation, say, via a change in the experimental conditions. In this spirit, we show in the main panel of Fig. 7, a variety in the $M_{FCC}\,(T)$ curves obtained at a chosen field of 50 Oe, when the amplitude of sample vibration in SVSM was changed from 0.5 mm to 8 mm. The following features in Fig. 7 are noteworthy:

1. The $M_{FCC}\,(T)$ curves at different amplitudes traverse different paths, which is very striking. In particular, the saturated values at the lowest temperature are significantly different during different runs (data shown only for the amplitudes of 0.5 mm and 8 mm in the inset of Fig. 7). Such a difference in saturated $M_{FCC}\,(T)$ is very similar to the observations made earlier for the $M_{FCC}(T)$ responses of the pristine and perturbed FC states (cf. inset panel of Fig. 3).

2. The paramagnetic peak can be seen to have highest value during the $M_{FCC}\,(T)$ run with an am-

plitude of 0.5 mm. The peak height was observed to steadily decrease as the amplitude was progressively increased from 0.5 mm to 7 mm.

3. At an amplitude of 8 mm, the $M_{FCC}\,(T)$ values can be seen to be diamagnetic in the entire temperature range $1.8\,\mathrm{K} < T < T_c$ (cf. the curve for 8 mm amplitude in the main panel and in the inset panel of Fig. 7).

Figure 7 thus illustrates a rich multiplicity in $M_{FCC}\,(T)$ getting exposed by the change in the amplitude of sample vibration. The similarity between the features emanating from $M_{FCC}\,(T)$ curves of inset panel in Fig. 7 and those of inset panel in Fig. 3 is curious.

It was observed earlier in Fig. 4(b) that an impulse, when applied above about 6.8 K, can switch a given diamagnetic value to a paramagnetic value (see perturbed (#1) in Fig. 4(b)). Along this line, it will now be interesting to investigate whether such a change can also be induced by an impulse in a $M_{FCC}\,(T)$ response at higher amplitude, say 8 mm, which in pristine conditions remained entirely diamagnetic below T_c (in Fig. 7). To demonstrate this, we applied an impulse on the pristine FC state created (at 8 mm amplitude) at $T = 7.05\,\mathrm{K}$ in a field of $H = 50\,\mathrm{Oe}$ as shown in Fig. 8(a). For comparison, we have appended in Fig. 8 the pristine $M_{FCC}\,(T)$ curve at 0.5 mm amplitude as well. It is interesting to note that the diamagnetic value of the pristine $M_{FCC}\,(T)$ curve for 8 mm amplitude changes to a paramagnetic value after the impulse treatment at $T = 7.05\,\mathrm{K}$. The $M_{FCC}\,(T)$ curve after perturbation (open triangles) was traced and found to be following a different path as shown in the main panel of Fig. 8(a). The saturated value of this curve (perturbed, 8 mm) at 2 K approaches nearer to that of pristine $M_{FCC}\,(T)$ curve at 0.5 mm as seen in the inset of Fig. 8(a). Similar investigations were carried out for the pristine $M_{FCC}(T)$ curve at 7 mm amplitude as shown in Fig. 8(b). Here, we performed two different runs (independently), after the impact of an impulse at $T = 7.05\,\mathrm{K}$. In one of the runs (perturbed (#1)), a paramagnetic magnetization was induced at 7.05 K and the $M_{FCC}\,(T)$ thereafter follows a certain path (open triangles), which is similar to that seen for 8 mm amplitude in the perturbed state (Fig. 8(a)). During the other run (perturbed (#2)), we observed that the magnetization data changes from near zero value of the pristine M_{FCC} for 7 mm to a large diamagnetic value after the imposition of an impulse at 7.05 K, and follows a path shown by open squares in Fig. 8(b). We draw an important inference here that the occurrence of paramagnetic response is not specific to a given amplitude of sample vibration, as it can be unearthed via a perturbation at all amplitudes ranging from 0.5 mm to 8 mm.

We finally exemplify in the main panel of Fig. 9 an unusual feature, viz., a characteristic oscillatory behaviour seen in $M_{FCC}\,(T)$ response recorded at 0.5 mm amplitude in a higher field, $H = 3\,\mathrm{kOe}$. This curve exhibits some peculiar undulations which override the paramag-

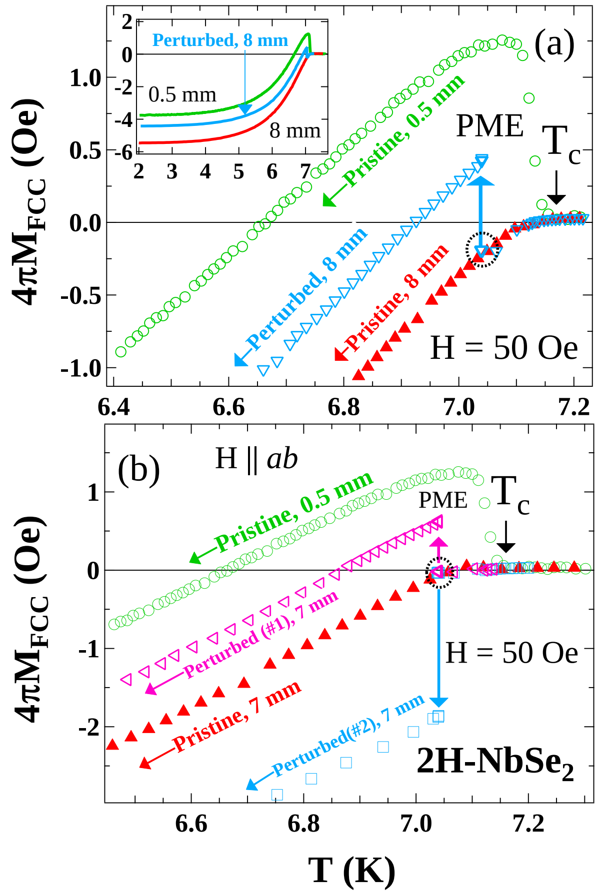

Figure 8: (Color online) (a) $M_{FCC}(T)$ in $H = 50\,\mathrm{Oe}$, recorded at amplitude of 0.5 mm and 8 mm, without imposing a perturbation during the cool-down are termed as pristine curves. The impulse treatment at 7.05 K switches a diamagnetic value of pristine M_{FCC} (8 mm) into a paramagnetic value, and the (perturbed, 8 mm) $M_{FCC}\,(T)$ curve was traced on further cooling. The inset panel in Fig. 8(a) displays the three curves in the range, $2\,\mathrm{K} < T < 7.5\,\mathrm{K}$. (b) $M_{FCC}\,(T)$ at $H = 50\,\mathrm{Oe}$ obtained at amplitudes of 0.5 mm and 7 mm, respectively in pristine conditions, plotted together with $M_{FCC}\,(T)$ obtained at 7 mm in the perturbed situations (#1 and #2). In perturbed (#1) state (open triangles), the $M_{FCC}\,(T)$ switches from near zero value to a paramagnetic value whereas in other case (#2), it switches into the diamagnetic region (open squares).

netic magnetization signal below T_c. It appears as if the magnetization shows a tendency to switch between the paramagnetic and diamagnetic responses while field-cooling. Such undulations echo similar oscillatory features reported earlier at lower field values ($< 14\,\mathrm{Oe}$) in another low T_c superconductor, viz., $Ca_3Rh_4Sn_{13}$ by Kulkarni *et al.* [24]. It may also be added here that the oscillatory behaviour in $M_{FCC}\,(T)$ is not merely restricted to lower amplitudes and higher fields, but, the same were also discernible at higher amplitudes and lower fields in 2H-$NbSe_2$. At 7 mm amplitude and in a field of

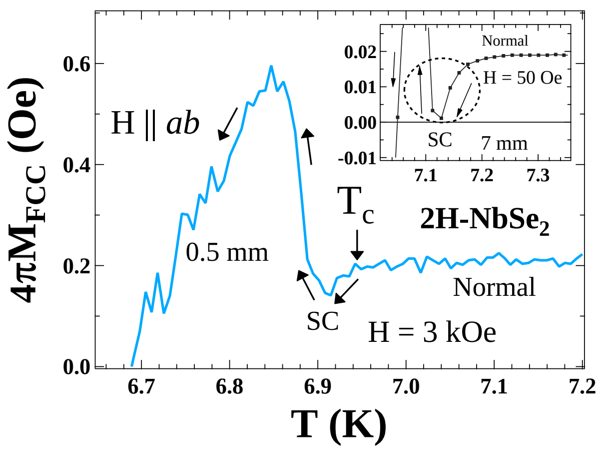

Figure 9: (Color online) A portion of the $M_{FCC}\,(T)$ curve at $H = 3\,\mathrm{kOe}$ recorded at an amplitude of 0.5 mm showing some unusual undulations. The inset panel shows a dip (see encircled portion) in $M_{FCC}\,(T)$ curve at $H = 50\,\mathrm{Oe}$, just below the onset of the superconductivity during run with 7 mm amplitude.

$H = 50\,\mathrm{Oe}$, the $M_{FCC}\,(T)$ response was observed to have an oscillatory character in close proximity of T_c as can be seen in an expanded portion of the $M_{FCC}\,(T)$ in an inset panel of Fig. 9. This curve first shows a pronounced dip (encircled in the inset of Fig. 9), which can be taken as the onset of diamagnetism. Following this dip, there occurs a sharp upturn on further lowering of temperature. The $M_{FCC}\,(T)$ values eventually cross over to negative values on lowering the temperature below $T \approx 7.05\,\mathrm{K}$.

IV. DISCUSSION

We have come across some amazing metastability effects in magnetization response on field-cooling a single-crystal of 2H-NbSe$_2$, viz., occurrence of both paramagnetic and diamagnetic response in a temperature window just below T_c, multi-valuedness in $M_{FCC}\,(T)$ at a given (H,T) value, oscillatory behaviour in a given $M_{FCC}\,(T)$ curve, unpredictable switching of $M_{FCC}\,(T)$ between diamagnetic and paramagnetic values due to an intervention by an external perturbation, etc. The mechanism and origin behind the said features in 2H-NbSe$_2$ cannot be comprehended on the basis of existing theoretical treatments in the literature. The following descriptions form the basis of this assertion:

1. The occurrence of (anomalous) paramagnetic magnetization in conventional bulk superconductors [13] had lead to several proposals, amongst which the flux-trapping model by Koshelev and Larkin [31] received significant attention. This model treats the situation wherein the magnetic flux gets trapped while field-cooling within a (macroscopic or mesoscopic) superconducting specimen due to an inhomogenous cooling [31]. While a (diamagnetic) current flows on the surface to shield an external field, associated with the trapped flux inside the specimen, there occurs a (paramagnetic) current that circulates in the interior of a superconducting sample [31]. Paramagnetic magnetization response can result due to an unbalancing effect of magnetization responses arising from the above stated two currents. The inhomogeneity in field-cooling, can have a variety, which in turn, can lead to a variety in trapped flux, thereby, resulting in multitude of magnetization responses during field-cooling experiments. Such a possibility could rationalize the observation of variety in $M_{FCC}\,(T)$ response, as reflected in Fig. 7 in the form of vibration amplitude-dependent $M_{FCC}\,(T)$ data at a given field value. Carrying this argument further, the data in Fig. 7 would imply that the inhomogeneity in cooling may be lesser at higher value of amplitude of vibration (see $M_{FCC}\,(T)$ curves at 7 mm and 8 mm amplitude in Figs. 7 and 8), leading to insignificant flux trapping (absence of paramagnetic signal). However, even in case of $M_{FCC}\,(T)$ data recorded at higher amplitudes (say 8 mm), that had remained entirely diamagnetic below T_c, a diamagnetic $M_{FCC}\,(T)$ value at a given temperature can be switched into a paramagnetic one, when the sample is perturbed by an impulse of an ac field (cf. Fig. 8(a)). From the perspective of flux-trapping model [31], this would imply that the impulse treatment can drive the system from a state of insignificant trapped-flux (diamagnetic) to a state of large trapped-flux (paramagnetic), which sounds to be unfeasible. Along similar lines, it can be argued that the occurrence of a diamagnetic dip just below T_c (insignificant trapped-flux) followed by a paramagnetic peak (large trapped-flux) in $M_{FCC}\,(T)$ (cf. Fig. 9) cannot be rationalized by this model. Also, the switching tendency of magnetization between diamagnetic and paramagnetic values identified via the characteristics undulations (oscillatory behaviour) in a given $M_{FCC}\,(T)$ run (Fig. 9) needs comprehension which is beyond the scope of flux-trapping model. We further noted two different effects of an impulse in two independent runs on the $M_{FCC}\,(T)$ response obtained at a higher amplitude of 7 mm (Fig. 8(b)). In one of the instances (perturbed (#1)), the impulse switches a diamagnetic (pristine) $M_{FCC}\,(T)$ value into a paramagnetic one while in the other (independent) run (#2), it switches the same diamagnetic $M_{FCC}\,(T)$ value into a larger diamagnetic value. Clearly, this suggests that the effects of an impulse in the present study are completely *random,* whereas the Koshelev and Larkin's mathematical treatment [31] to a given field and current distribution due to a trapped flux ought to be *predictable.* We therefore rule out that this model can explain the metastability observed in $M_{FCC}\,(T)$ response of 2H-NbSe$_2$.

2. To understand the thermomagnetic history dependence observed in the magnetization data of a type-II superconductor, Clem and Hao [42] had constructed a model which relies on the Bean's Critical State framework [43]. Although this model [42] yields the flux-

density profiles that can get set up in a superconductor during different protocols usually studied by experimentalists, viz., zero field-cooled, field-cooled cool-down and field-cooled warm-up modes, it cannot account for paramagnetic magnetization in the superconducting specimen. However, we noted that the effect of an impulse applied below $T = 6.7\,\mathrm{K}$ does not lead to any paramagnetic magnetization but an enhanced diamagnetism (cf. Fig. 5). The multi-valuedness in (diamagnetic) $M_{FCC}(T)$ data at low temperatures ($T < 6.7\,\mathrm{K}$, cf. inset panel of Fig. 3, 4(a) and 7 and 8(a) and Fig. 5) can be related in an oblique way to a result that emanated using the model of Clem and Hao [42]. We recall that the flux expulsion by a pinned-superconductor when it is field-cooled through its T_c is never complete [42] and therefore the field-cooled magnetization generally does not conform to an equilibrium magnetization. The model by Clem and Hao [42] indeed shows that the $M_{FCC}(T)$ curve at a constant field depends on the pinning strength; a more (less) diamagnetic state amounts to less (more) strongly pinned vortex configuration (see Fig. 5 in Ref. [42]). We observe a diversity in $M_{FCC}(T)$ data (see, for example, inset of Fig. 3) that closely resembles the pinning-dependent (multi-valued) $M_{FCC}(T)$ curves numerically calculated by Clem and Hao (see Fig. 5 in Ref. [42]), however, the multiplicity seen in present case is in the same 2H-$NbSe_2$ crystal, i.e., without changing the amount of quenched-disorder (pinning strength). Comparing the two pictures, viz., the $M_{FCC}(T)$ response of 2H-$NbSe_2$ (an inset of Fig. 3) and that shown in Fig. 5 in Ref. [42], one may naively surmise that the non-uniqueness in $M_{FCC}(T)$ in 2H-$NbSe_2$ could be due to the possibility of coexistence of several (metastable) vortex configurations with varying pinning strength. The impulse treatment or amplitude-variation can lead to the switching of the system from one configuration to another, which is reflected by the diversity in $M_{FCC}(T)$ as shown in inset panels of Figs. 3 and 7. In a given field-cooled run, the $M_{FCC}(T)$ values get saturated below a certain temperature, T_{c1}, at which the applied field just equals the lower critical field ($H_{c1}(T)$). Below T_{c1}, i.e., in the Meissner phase, the applied field remains smaller than the $H_{c1}(T)$ value and the application of an ac field impulse (h_{ac}) superimposed on the (static) applied field ($H \pm h_{ac}$) is hardly expected to produce any significant change in $M_{FCC}(T)$. This is apparent from the minimal changes seen in $M_{FCC}(T)$ below $T = 5\,\mathrm{K}$ for the perturbed cases (#2), (#3) and (#4) in Fig. 5.

3. 2H-$NbSe_2$ system has for long remained a favourite compound to explore novel notions in vortex state studies. In particular, this compound has been very widely investigated (see Ref. [44] and references therein) for order-disorder transitions as fingerprinted via anomalous variations in field/temperature dependencies in critical current density, viz., peak effect (PE) phenomenon/second magnetization peak (SMP) anomaly in magnetization hysteresis loops. Due to supercooling/superheating effects [45] that can occur across these transitions, the magnetization response is generally found to be history-dependent [44]. However, we would like to emphasize here that the present results pertaining to metastability effects in 2H-$NbSe_2$ have no correlations with the metastability and thermomagnetic history-dependence in critical current density values seen across the PE/SMP transitions. Firstly, most of the magnetization data presented here have been recorded at very low fields (i.e., $H = 50\,\mathrm{Oe}$). We have checked that at this field value, the present crystal does not display either PE in temperature-dependent isofield scans or SMP transition in isothermal scans at any temperature. Secondly, the history-dependent magnetization across the PE/SMP has never been reported to generate paramagnetic magnetization.

4. Clearly, all the novel proposals [4, 8, 9, 27–30] that have been put forward to understand the origin of paramagnetic magnetization in high-T_c superconductors are very difficult to visualize in the present context which involves a conventional low-T_c superconductor. Even the controlled manipulation of paramagnetic magnetization demonstrated recently [26] in Pb-Co nanocomposites involved the change in the orientation of magnetic moments of Co nanoparticles unlike the present situation wherein we have observed in a non magnetic superconductor, 2H-$NbSe_2$, an unpredictable switching of paramagnetic magnetization into diamagnetic and vice versa.

5. We rule out that the present results are a consequence of any specific disorder present in the anisotropic 2H-$NbSe_2$ crystal as there have been few other reports of similar metastability effects (including oscillatory magnetization behaviour) in other isotropic low-T_c superconductors, viz., single-crystals of $Ca_3Rh_4Sn_{13}$ [24] and Nb (in the form of a sphere) [18]. An advantage with a spherical (Nb) crystal [18] is that the demagnetization factor remains the same irrespective of the sample orientation with respect to field and, hence, yields identical results in all orientations. In these studies [18, 24], however, the effects of perturbation on $M_{FCC}(T)$ response were not explored. The novel consequences of perturbation on the $M_{FCC}(T)$ response in the present study have lead to exposure of a rich multiplicity in FC magnetization and random switching tendency of $M_{FCC}(T)$ between paramagnetic and diamagnetic values, which is indeed an advancement over the results reported in Ref. [18, 24].

We now focus our attention onto some theoretical works [32, 33, 36–40] which attempt to account for the occurrence of PME in mesoscopic-sized samples. Based on the Ginzburg-Landau theory, the theoretical pictures framed exclusively for mesoscopic superconductors foresee the occurrence of giant vortex states with multiple flux quanta ($L > 1$) at the onset of surface superconductivity [34]. Each multi-quantum state has its own field domain of existence; at higher field end of each domain, the response is diamagnetic, which crosses over to the paramagnetic values as the field decreases (see Fig. 2(b) in Ref. [33], Figs. 16, 17 and 23 in Ref. [40]). It is also possible that the temperature (or field) sweeping in a given

$M(T)$ (or M–H) run is sufficient enough to drive the system away from the field/temperature domain of a certain configuration (L), which may induce a forced transition [33, 40] to the nearest minimum energy configuration (with different L value). An examination of the M–H plots for different multi-quanta states ($L > 1$) along with the corresponding free energy vs field curves drawn for a specific circumstance in Figs. 2 and 3 of Ref. [33] tells us that if the transitions occur between L states such that the configuration pertaining to a certain L value with the lowest free energy prevails, there would be no metastability effects in the magnetization response and the magnetization values as a function of temperature (in isofield scans) would be diamagnetic all through. However, if metastability is permissible and transitions happen between different metastable L states, one can encounter all sorts of possibilities, i.e., the magnetization values can change from a given paramagnetic/diamagnetic value to a higher or a lower value, and it can also transit from positive to negative values or vice versa.

To our knowledge, the above mentioned (and other related) studies [32, 33, 36–40] describing the magnetization behaviour of multi-quanta states ($L > 1$) in mesoscopic samples are the only examples which bear some resemblance with our findings. We draw here some parallels between these theoretical findings [32, 33, 36–40] and our experimental results in a single crystal of 2H-$NbSe_2$, to which the latter results [32, 33, 36–40] prima facie do not apply. As mentioned above, each multi-quantum state ($L > 1$) has a certain domain of its existence wherein the magnetization response can be either diamagnetic or paramagnetic depending upon the external field [33]. Similarly, in 2H-$NbSe_2$, there exists a temperature window ($6.7\,\mathrm{K} < T < T_c$), where a perturbation applied to the system (at a given (H,T) value) can randomly yield the magnetization which is either paramagnetic or diamagnetic (see Figs. 3, 4(b), 6 and 8). The switching of $M_{FCC}(T)$ due to a perturbation leads to a rich diversity in magnetization response in 2H-$NbSe_2$ (cf. Figs. 3 to 6 and 8), very similar to characteristics of magnetization behaviour anticipated during the transformation amongst various multi-quanta ($L > 1$) states [33, 40]. Additionally, the oscillatory behavior seen in $M_{FCC}(T)$ curve (Fig. 9) mimics the magnetization response predicted [33, 40] for the transformations amongst multi-quanta states ($L > 1$) during the temperature-sweep. In an earlier unrelated study, the scanning tunneling microscopy technique employed by Karapetrov *et al.* [46], enabled them to directly observe a coexistence of multi-quanta vortex states and Abrikosov lattice in an array of submicroscopic metal (gold) island embedded in a single crystal of 2H-$NbSe_2$. The simultaneous existence of multi-quanta state of different vorticities is thus not an unrealistic possibility.

In spite of a strong resemblance of our results with the characteristics of multi-quanta states [32, 33, 36–40]*, the possibility of occurrence of multi-quanta states in the sample investigated here remains a speculation considering the larger size of this (macroscopic) sample. The effect of multi-vorticity in such a macroscopic sample is an exotic proposition. Therefore, there is currently a necessity to look for a more sophisticated model to fully understand the new findings in 2H-$NbSe_2$ as well as earlier results in $Ca_3Rh_4Sn_{13}$ [24] and Nb [18].*

In the end, we rule out the possibility of any experimental artefact arising due to change in amplitude of vibration considering the following important points: (i) The factor which could be affected most by changing the sample vibration is the magnetic field-inhomogeneity which, in the present case, is found to be negligible ($\sim 10^{-2}$ Oe). (ii) We did not register any change in the magnetization on varying the amplitude at a given (H,T) value in the case of a standard Indium sample. (iii) We have also shown that the PME like attribute in 2H-$NbSe_2$ is not limited to only smaller amplitudes but the same can also be induced at higher amplitudes (8 mm), after the impulse treatment.

V. CONCLUSION

We have investigated in detail a single crystal specimen of 2H-$NbSe_2$ via dc magnetization measurements. The new revelations in this compound correspond to the observation of an anomalous paramagnetic (PME like) signal(s) below T_c and some peculiar consequences of applying a perturbation at various temperatures on a given field-cooled state. Based on the effects of perturbation, two distinct temperature intervals have been identified. Across a certain temperature range lying just below T_c, the PME-like signal can be manipulated by applying an external perturbation, which unpredictably switches the $M_{FCC}(T)$ response from a given para/diamagnetic value into a different para/diamagnetic value. On the other hand, when the system is perturbed away from T_c (below about $T = 6.7\,\mathrm{K}$), the effect of perturbation is of one kind, i.e., it only enhances the diamagnetism such that no paramagnetic signal is seen. The saturated (diamagnetic) $M_{FCC}(T)$ value at the lowest temperature is found to be influenced by the effect of perturbation applied closer to the region of PME. The said features in $M_{FCC}(T)$ data in a single crystal of 2H-$NbSe_2$ coincidentally bear similarities with the magnetization response predicted [32, 33, 36–40] for mesoscopic samples, wherein, a possibility of nucleation of multi-quanta states ($L > 1$) have been discussed in the literature. We believe the present results in a bulk 2H-$NbSe_2$ crystal cannot be explained by any of the existing mathematical treatments based on prescriptions of Bean's Critical State model [43] reported in the literature and call for a new theoretical framework. Though, we have presented experimental results pertaining to a given orientation of magnetic field vis. a vis. crystallographic direction ($H \,||\, ab$) of hexagonal 2H-$NbSe_2$, we have also carried out magnetization measurements in more often studied orientation, viz., $H \,||\, c$, in 2H-$NbSe_2$ and obtained nearly identical features (data not included in the present report). We may therefore state that assertions made in this report are not specific to the anisotropic nature of the intrinsic superconducting parameters of 2H-$NbSe_2$.

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