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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 magnetic 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 ($L\Phi_0, \Phi_0 = \hbar c/2e, L > 1$) predicted to occur in mesoscopic-sized superconducting specimen and possible transitions amongst such states.

Keywords: field-cooled magnetization response in superconductors, paramagnetic magnetization response, metastability effects

(Some figures may appear in colour only in the online journal)

1. 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 [27] for a review), e.g. d-wave superconductivity [9], orbital glass [27, 28], presence of $\pi$ 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 a multi-quanta vortex matter, i.e. vortices with multiple flux quanta, \( L \Phi_0 \), \( \Phi_0 = \hbar c/2 e \), \( L > 1 \), nucleating below a third critical field \( (H_c^3) \), 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. This compound has been in focus in recent times for studies pertaining to charge density wave transition [36–38]. The results being reported here for 2H-NbSe\(_2\) present an advancement over the studies reported earlier by some of us in Ca\(_3\)Rh\(_2\)Sn\(_3\) [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 metastable multiple flux quanta states \( (L > 1) \) proposed in the context of mesoscopic samples [32, 33, 39–43].

2. Experimental details

The single crystals of 2H-NbSe\(_2\) were grown by Iodine vapor transport method [44]. The sample chosen for the present study is platelet-shaped single crystal with a planar area of 4.12 mm\(^2\), thickness of 0.17 mm and mass of 4.64 mg. It belongs to the same batch of 2H-NbSe\(_2\) crystals as studied by Ghosh et al, [45] and Banerjee et al, [46]. The superconducting transition temperature of the present sample is found to be \( \approx 7.15 \) K, which indicates that this sample is comparable to the sample Y \((T_c(0)\approx 7.17 \) K) utilized by Banerjee et al, [46]. The 2H-NbSe\(_2\) crystals grown by vapor transport method are typically very clean and exhibit very low critical current density. 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. \( H \parallel ab \)), 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 \((-10^{-1})\) and the associated geometrical/boundary effects are expected to be minimal. We have 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 [47] 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 figure 7), which is argued to be a consequence of the metastability effects prevailing in this system, a key focus of the present report.

3. Results

3.1. 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 (\( M_{FC}(T) \)) 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 magnetic field. A field, \( H = 50 \) Oe, was then applied (such that \( H \parallel ab \)) and the magnetization was 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 \( (M_{ZFC}(T)) \). Thereafter, the magnetization values were measured again while warming, to yield \( M_{FCW}(T) \). The \( M_{ZFC}(T) \) curve shows the usual fall in diamagnetic response (see inset panel of figure 1(a)) of a superconducting specimen as the temperature is swept up.
The multi-valuedness in $M(T)$ observed in figure 1(b) motivated us to explore further the magnetization responses in 2H-NbSe$_2$. We illustrate in the main panel of figure 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 figure 3) while field-cooling the sample to 2 K, similar to the way $M_{FCC}(T)$ data of figure 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 the peak temperature ($T_p$) of the PME, i.e. $T \approx 7.07$ K, which is encircled in the main panel of figure 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 s. We have verified that the results reported in figure 3 do not depend on the duration of this 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 the main panel of...
Figure 3. $M_{\text{FCC}}(T)$ 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 $M_{\text{FCC}}(T)$ traverses different paths as displayed for three different runs, viz., perturbed (#1), perturbed (#2) and perturbed (#3). The pristine $M_{\text{FCC}}(T)$ (open circles) was recorded without perturbing the system throughout the cooling. The inset displays $M_{\text{FCC}}(T)$ for pristine and perturbed (#1)–(#3) cases in the full range, $2 \text{ K} < T < 7.5 \text{ K}$.

Figure 4. $M_{\text{FCC}}(T)$ 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 $M_{\text{FCC}}(T)$. 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) $M_{\text{FCC}}(T)$ 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.

Subsequent efforts of applying the impulse not only at $T_p$ but also in its vicinity, viz., across $6.75 \text{ K} < T < T_c$ (all data not shown here), lead to the inference that the $M_{\text{FCC}}(T)$ value, after perturbation in the PME region, can unproductively change very widely from paramagnetic to diamagnetic or from a given paramagnetic to a less/more paramagnetic value. We have also applied impulses with different amplitudes and 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 figure 3. When subjected to a perturbation, the system can transit between these metastable states, as is reflected from the abrupt change(s) in magnetization.

3.3. Path dependent $M(T)$ response

We now demonstrate in figure 4, the impact of an ac field impulse applied at various temperatures, during a given
$M_{\text{FCC}} (T)$ run at $H = 50 \, \text{Oe}$. The pristine $M_{\text{FCC}} (T)$ shown in figure 4 is the same as that presented in figure 3. When an impulse was applied at $T = 7 \, \text{K}$ (encircled in figure 4(a)), an enhanced paramagnetic signal was seen as depicted by the $M_{\text{FCC}} (T)$ curve named ‘perturbed (#1)’. This curve crosses over to the diamagnetic region smoothly at $T \sim 6.4 \, \text{K}$, and thereafter remains less diamagnetic than the pristine $M_{\text{FCC}} (T)$ until about $T = 6 \, \text{K}$, echoing the ‘perturbed (#2)’ case of figure 3. At $T = 6 \, \text{K}$, an impulse was imposed again (see the upper encircling done corresponding to 6 K in figure 4(a)) and the $M_{\text{FCC}}$ 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 figure 4(a), which is different from its pristine counterpart. The saturated $M_{\text{FCC}} (T)$ value at 2 K corresponding to perturbed (#1) curve is found to be nearly twice the saturated $M_{\text{FCC}} (T)$ in the pristine case, as is evident in the inset panel of figure 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 figure 4(a)). Such an impulse treatment resulted in the diamagnetic value of pristine $M_{\text{FCC}} (T)$ (encircled) which also comprised impulse treatment(s) at several intermediate temperatures (see the steps in magnetization where the impulse was applied) during the cool-down process. The results in figures 4 and 5 focus on $M_{\text{FCC}} (T)$ data, while subjecting the sample to perturbations at intermediate temperatures during cool-down cycle. In figure 6, we show another
Figure 6. $M_{\text{FCC}}(T)$ curves at $H = 50$ Oe traced in five different warm-up runs after perturbation by an ac field impulse (perturbed (#1)–(#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)–(#5) were recorded while warming the sample to higher $T$, after perturbing the pristine FC state created at $T = 7$ K (perturbed (#1) and perturbed (#2)), $T = 6.71$ K (perturbed (#3)) and $T = 7.1$ K (inset shows perturbed (#4) and (#5)). In all the five cases, the $M_{\text{FCC}}(T)$ curves show a variety of paths that can be traversed while warming after the perturbation.

Figure 7. Portions of the $M_{\text{FCC}}(T)$ curves recorded in $H = 50$ Oe at various amplitudes of vibration in a VSM, as indicated. The saturated values of $M_{\text{FCC}}$ at $T \sim 1.8$ 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.

3.4. Diversity in $M_{\text{FCC}}$ 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 (figures 3–6). It is also tempting to ask whether a multiplicity in $M(T)$ similar to that observed in figures 3–6 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 figure 7, a variety in the $M_{\text{FCC}}(T)$ 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 figure 7 are noteworthy:

1. The $M_{\text{FCC}}(T)$ 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 figure 7). Such a difference in saturated $M_{\text{FCC}}(T)$ is very similar to the observations made earlier for the $M_{\text{FCC}}(T)$ responses of the pristine and perturbed FC states (see inset panel of figure 3).

2. The paramagnetic peak can be seen to have the highest value during the $M_{\text{FCC}}(T)$ run with an amplitude 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 $M_{\text{FCC}}(T)$ values can be seen to be diamagnetic in the entire temperature range 1.8 K $< T < T_c$ (see the curve for 8 mm amplitude in the main panel and in the inset panel of figure 7).

Figure 7 thus illustrates a rich multiplicity in $M_{\text{FCC}}(T)$ getting exposed by the change in the amplitude of sample...
Figure 8. (a) $M_{FCC}(T)$ in $H = 50$ 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 figure 8(a) displays the three curves in the range, 2 K $< T < 7.5$ K. (b) $M_{FCC}(T)$ at $H = 50$ 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).

The similarity between the features emanating from the $M_{FCC}(T)$ curves in the inset panel in figure 7 and those from the inset panel in figure 3 is curious.

It was observed earlier in figure 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 figure 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 figure 7). To demonstrate this, we applied an impulse on the pristine FC state created (at 8 mm amplitude) at $T = 7.05$ K in a field of $H = 50$ Oe as shown in figure 8(a). For comparison, we have appended in figure 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 $T = 7.05$ 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 figure 8(a). The saturated value of this curve (perturbed, 8 mm) at 2 K approaches nearer to that of the pristine $M_{FCC}(T)$ curve at 0.5 mm as seen in the inset of figure 8(a). Similar investigations were carried out for the pristine $M_{FCC}(T)$ curve at 7 mm amplitude as shown in figure 8(b). Here, we performed two different runs (independently), after the impact of an impulse at $T = 7.05$ 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 (figure 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 figure 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 figure 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, $H = 3$ kOe. This curve exhibits some peculiar undulations which override the paramagnetic 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 Oe) in another low $T_c$ superconductor, viz., $CaRh_5Sn_{13}$ 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$_2$. 

![Figure 9](image-url)
At 7 mm amplitude and in a field of $H = 50$ Oe, the $M_{\text{FCC}}(T)$ response was observed to have an oscillatory character in close proximity of $T_c$ as can be seen in an expanded portion of the $M_{\text{FCC}}(T)$ in an inset panel of figure 9. This curve first shows a pronounced dip (encircled in the inset of figure 9), which can be taken as the onset of diamagnetism. Following this dip, there occurs a sharp upturn on further lowering of temperature. The $M_{\text{FCC}}(T)$ values eventually cross over to negative values on lowering the temperature below $T \approx 7.05$ K.

4. Discussion

We have come across some amazing metastability effects in the 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 $M_{\text{FCC}}(T)$ at a given $(H,T)$ value, oscillatory behaviour in a given $M_{\text{FCC}}(T)$ curve, unpredictable switching of $M_{\text{FCC}}(T)$ 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 inhomogeneous cooling [31]. While a (diamagnetic) current flows on the surface to shield an (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 two above stated 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 the field-cooling experiments. Such a possibility could rationalize the observation of variety in $M_{\text{FCC}}(T)$ response, as reflected in figure 7 in the form of vibration amplitude-dependent $M_{\text{FCC}}(T)$ data at a given field value. Carrying this argument further, the data in figure 7 would imply that the inhomogeneity in cooling may be lesser at a higher value of amplitude of vibration (see $M_{\text{FCC}}(T)$ curves at 7 mm and 8 mm amplitude in figures 7 and 8), leading to insignificant flux trapping (absence of paramagnetic signal). However, even in the case of $M_{\text{FCC}}(T)$ data recorded at higher amplitudes (say 8 mm), that had remained entirely diamagnetic below $T_c$, a diamagnetic $M_{\text{FCC}}(T)$ value at a given temperature can be switched into a paramagnetic one, when the sample is perturbed by an impulse of an ac field (see figure 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 $M_{\text{FCC}}(T)$ (see figure 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 $M_{\text{FCC}}(T)$ run (figure 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 $M_{\text{FCC}}(T)$ response obtained at a higher amplitude of 7 mm (figure 8(b)). In one of the instances (perturbed (1)), the impulse switches a diamagnetic (pristine) $M_{\text{FCC}}(T)$ value into a paramagnetic one while in the other (independent) run (2), it switches the same diamagnetic $M_{\text{FCC}}(T)$ 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 the $M_{\text{FCC}}(T)$ responses of 2H-NbSe$_2$.

2. To understand the thermomagnetic history dependence observed in the magnetization data of a type-II superconductor, Clem and Hao [48] had constructed a model which relies on the Bean’s Critical State framework [49]. Although this model [48] 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 $T = 6.7$ K does not lead to any paramagnetic magnetization but an enhanced diamagnetism (see figure 5). The multi-valuedness in (diamagnetic) $M_{\text{FCC}}(T)$ data at low temperatures ($T < 6.7$ K, see inset panel of figures 3, 4(a), 5, 7 and 8(a)) can be related in an oblique way to a result that emanated using the model of Clem and Hao [48]. We recall that the flux expulsion by a pinned-superconductor when it is field-cooled through its $T_c$ is never complete [48] and therefore the field-cooled magnetization generally does not conform to an equilibrium magnetization. The model by Clem and Hao [48] indeed shows that the $M_{\text{FCC}}(T)$ curve at a constant field depends on the pinning strength; a more (less) diamagnetic state amounts to less (more) strongly pinned vortex configuration (see figure 5 in [48]). We observe a diversity in $M_{\text{FCC}}(T)$ data (see, for example, inset of figure 3) that closely resembles the pinning-dependent (multi-valued) $M_{\text{FCC}}(T)$ curves numerically calculated by Clem and Hao (see figure 5 in [48]), 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 $M_{\text{FCC}}(T)$ response of 2H-NbSe$_2$ (an inset of figure 3) and that shown in figure 5 in [48], one may naively surmise that the non-uniqueness in $M_{\text{FCC}}(T)$ 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 $M_{\text{FCC}}(T)$ as shown in the inset panels of figures 3 and 7. In a given field-cooled run, the $M_{\text{FCC}}(T)$ values get saturated below a certain temperature, $T_{\text{c}1}$, at which the applied field just equals the lower critical field ($H_{\text{c}1}(T)$). Below $T_{\text{c}1}$, i.e. in the Meissner phase, the applied field remains smaller than the $H_{\text{c}1}(T)$ value and the application of an ac field impulse ($h_{\text{ac}}$) superimposed on the (static) applied field ($H \pm h_{\text{ac}}$) is hardly expected to produce any significant change in $M_{\text{FCC}}(T)$. This is apparent from the minimal changes seen in $M_{\text{FCC}}(T)$ below $T = 5$ K for the perturbed cases (#2)–(#4) in figure 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 [46] and references therein) for order-disorder transitions as fingerprinted via anomalous variations in field/temperature dependencies in critical current density, viz., the peak effect (PE) phenomenon/second magnetization peak (SMP) anomaly in magnetization hysteresis loops. Due to supercooling/superheating effects [50] that can occur across these transitions, the magnetization response is generally found to be history-dependent [46]. 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 the 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. $H = 50$ 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 the magnetic moments of Co nanoparticles unlike in 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 the possibility 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$_3$Rh$_2$Sn$_3$ [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 $M_{\text{FCC}}(T)$ response were not explored. The novel consequences of perturbation on the $M_{\text{FCC}}(T)$ response in the present study have lead to the exposure of a rich multiplicity in the FC magnetization and a random switching tendency of $M_{\text{FCC}}(T)$ between the paramagnetic and diamagnetic values, which is indeed an advancement over the results reported in [18, 24].

We now focus our attention onto some theoretical works [32, 33, 39–43] 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 the 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 figure 2(b) in [33], figures 16, 17 and 23 in [43]). 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, 43] to the nearest minimum energy configuration (with different $L$ value). An examination of the $M–H$ plots for different multi-quantum states ($L > 1$) along with the corresponding free energy versus field curves drawn for a specific circumstance in figures 2 and 3 of [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, 39–43] 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, 39–43] and our experimental results in a single crystal of 2H-NbSe$_2$, to which the latter results...
due to a value at data in a single crystal of 2H-

curve (figure 9) mimics the magnetization response
diamagnetic or paramagnetic depending upon the external
existence wherein the magnetization response can be either
characteristics of multi-quanta states
simultaneous existence of multi-quanta state of different vor-
(gold) island embedded in a single crystal of 2H-
magnetic field vis. a vis. crystallographic direction (H || ab) of
hexagonal 2H-NbSe₂ we have also carried out magnetization
measurements in more often studied orientation, viz., H || c,
in 2H-NbSe₂ 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 aniso-
tropic nature of the intrinsic superconducting parameters of
2H-NbSe₂.

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In spite of a strong resemblance of our results with the characteristics of multi-quanta states [32, 33, 39–43] , 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 multivorticity 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-NbSe₂ as well as earlier results in Ca₃Rh₄Sn₃ [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 (∼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-NbSe₂ is not limited to only smaller amplitudes but the same can also be induced at higher amplitudes (8 mm), after the impulse treatment.

5. Conclusion

We have investigated in detail a single crystal specimen of 2H-NbSe₂ via dc magnetization measurements. The new revelations in this compound correspond to the observation of an anomalous paramagnetic (PME like) signal(s) below Tc 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 Tc, 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 Tc (below about T = 6.7 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-NbSe₂ coincidentally bear similarities with the magnetization response predicted [32, 33, 39–43] 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-NbSe₂ crystal cannot be explained by any of the existing mathematical treatments based on prescriptions of Bean’s Critical State model [49] 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-NbSe₂, we have also discovered magnetization measurements in more often studied orientation, viz., H || c, in 2H-NbSe₂ 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-NbSe₂.
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