

Multiple magnetization peaks in weakly pinned $\text{Ca}_3\text{Rh}_4\text{Sn}_{13}$ and $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$

S. Sarkar,¹ D. Pal,¹ P. L. Paulose,¹ S. Ramakrishnan,^{1,*} A. K. Grover,^{1,*} C. V. Tomy,² D. Dasgupta,³ Bimal K. Sarma,³ G. Balakrishnan,⁴ and D. McK. Paul⁴

¹*Department of Condensed Matter Physics and Materials Science, Tata Institute of Fundamental Research, Mumbai 400005, India*

²*Department of Physics, Indian Institute of Technology, Powai, Mumbai 400 076, India*

³*Department of Physics, University of Wisconsin, Milwaukee, Wisconsin 53201*

⁴*Department of Physics, University of Warwick, Coventry CV4 7AL, United Kingdom*

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The second magnetization peak and the peak effect anomaly coexisting in a given isothermal magnetization hysteresis loop show striking similarities in $\text{Ca}_3\text{Rh}_4\text{Sn}_{13}$, a low- T_c superconductor and $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, a high- T_c superconductor. The observed variation of the hysteresis width with field could imply a modulation in the degree of the plastic deformation of the elastic vortex solid. The characteristics of the high- T_c cuprates, such as large Ginzburg number, short coherence length, decoupling of the Josephson coupled pancake vortices, etc., are unlikely to be the cause of the observed behavior.

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The inhomogeneities in the atomic lattice are convenient pinning sites for the normal cores of the vortex lines and this localization leads to a threshold critical current density J_c for the superconductor. Hence, J_c is a material attribute and it could provide information on the state of the spatial and temporal correlations in the vortex matter. The collective pinning description of the vortex medium due to Larkin-Ovchinnikov¹ relates J_c inversely to the correlation volume V_c (i.e., $J_c \propto V_c^{-1/2}$), within which the vortex array responds elastically, while nominally retaining the translation symmetry of the flux line lattice (FLL). The deviations from the usual monotonic decay in $J_c(H, T)$ with increase in the field (H) or the temperature (T) have been reported to span different regions of the thermomagnetic (H, T) phase space in a variety of superconductors since the early 1960's.²⁻¹¹ Isothermal M - H loops conveniently provide the information on $J_c(H)$ through the width of the hysteresis loop, $\Delta M(H) [=M(H_\uparrow) - M(H_\downarrow)]$, where $M(H_\uparrow)$ and $M(H_\downarrow)$ represent the magnetization in the increasing and decreasing directions of the magnetic field, respectively]. The two well documented anomalous behavior in $\Delta M(H)$ (or J_c) are the fish-tail effect (FE) (also often referred to as the second magnetization peak) and the peak effect (PE). The FE derives its name from the characteristic shape of the M - H loop and also corresponds to a hump feature in $J_c(H)$ far below the upper critical field H_{c2} . The PE, as the name may suggest, is usually identified with a well-defined peak in $J_c(H, T)$ on approaching the H_{c2} boundary. The PE phenomenon, ever since the initial proposal by Pippard,¹² is widely believed to be signaling a rapid collapse of the elastic moduli of the vortex solid vis a vis that of the elementary pinning force at the incipient FLL melting transition. On the other hand, for the so-called second magnetization peak (SMP) which is ubiquitous in high- T_c cuprates, a variety of explanations have appeared in the recent literature. These range from (i) the enhancement in pinning due to matching effects in oxygen deficient structures,¹³ (ii) the collective creep phenomenon,¹⁴ (iii) the surface barrier effect,¹⁵ (iv) the ther-

momagnetic instability,¹⁶ (v) the nonuniform current flow,¹⁷ (vi) the interplay between disordered and ordered regions,¹⁸ (vii) the Bragg glass (BG) (dislocation free vortex solid) to a vortex glass (VG) transition due to proliferation of dislocations,¹⁹ etc. Thus, the SMP and the PE are being discussed in apparently different terms. Recent simulation studies²⁰ of the driven vortex matter also anticipate two anomalous maxima in $J_c(H)$ at the interfaces of the Bragg glass to the vortex glass and the vortex glass to vortex liquid transitions. The issue of possible connection and distinction between the SMP and the PE remains open experimentally⁴⁻⁷ and a comprehensive theoretical account for both the effects is still lacking. Some of us^{10,21} have recently reported the observation of the splitting of the composite fish-tail-effect-like behavior in the M - H loop of $2H$ -NbSe₂ ($T_c \approx 7.2$ K) into two well separated anomalies in $J_c(H)$, one of which could be termed as the plateau effect and the other as the usual PE. The plateau effect relates to a crossover [or transition^{10,21,22}] in a characteristic manner from an interaction-dominated collectively pinned state to a small bundle (or individual) pinning regime at low fields where the inter vortex spacing a_0 ($\propto 1/\sqrt{B}$) exceeds the range of the interaction λ (i.e., the penetration depth) between the vortices. In this paper, we report the occurrence of the SMP and the PE and their striking resemblances in single crystals of two entirely different types of superconductors $\text{Ca}_3\text{Rh}_4\text{Sn}_{13}$ (CaRhSn), an isotropic low- T_c superconductor ($T_c \approx 8.2$ K)²³ and $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO), an anisotropic high- T_c cuprate superconductor [$T_c \approx 93.2$ K, $\Delta T_c = 0.8$ K]. The YBCO crystal is the same piece, with a very low density of twin boundaries,²⁴ in which the locus of the PE temperatures $T_p(H)$ displays a reentrant characteristic at low H , analogous to the behavior first reported in $2H$ -NbSe₂ (Ref. 25) and identified with the reentrant nature of the ideal (pinning free) FLL melting curve.²⁶

The present results in CaRhSn and YBCO, in conjunction with the earlier data from $2H$ -NbSe₂ (Ref. 21) and YBCO,⁴⁻⁶ not only establish the distinct character of the SMP and the PE, but, also, inter alia illustrate the different

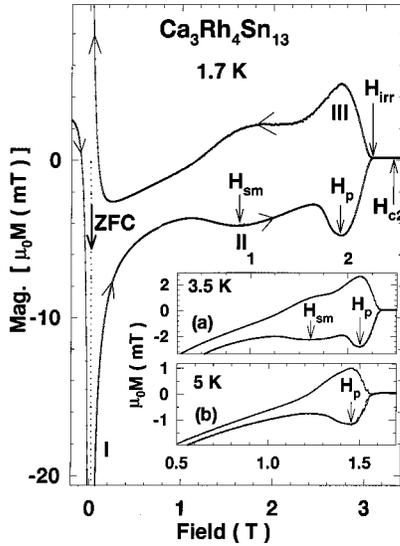


FIG. 1. M - H loop in a single crystal of $\text{Ca}_3\text{Rh}_4\text{Sn}_{13}$ at 1.7 K. The three peaks in the magnetization are marked as I, II, and III. The forward leg ($-H_{\text{max}}$ to $+H_{\text{max}}$) of the envelope loop meets the virgin ZFC M - H curve just above the peak I at $H \sim 17$ mT. H_{sm} and H_{p} mark the maxima of the SMP and the PE, respectively. H_{irr} and $H_{\text{c}2}$ have also been identified at 1.7 K. The insets (a) and (b) show portions of the M - H loops at 3.5 and 5 K, respectively.

circumstances in which the two either overlap or one completely dominates the other. Further, the observation of the similar features in both a low- T_c and a high- T_c superconductor points²⁷ towards the existence of a characteristic behavior for the vortex matter, notwithstanding (i) the wide differences^{9,28} in the values of the Ginzburg number G_i $\{=[1/2][k_B T_c / H_c^2(0) \epsilon \xi^3(0)]^2\}$ of these two different classes of superconductors and (ii) the additional complexity (arising out of the Josephson coupling) in the constitution of an individual vortex line in the high- T_c cuprates.²⁸

The M - H loops in CaRhSn ($H \parallel \text{cube edge}$) and YBCO ($H \parallel c$) were recorded using a vibrating sample magnetometer (Oxford Instruments, U.K.). Figures 1 and 2 display data for

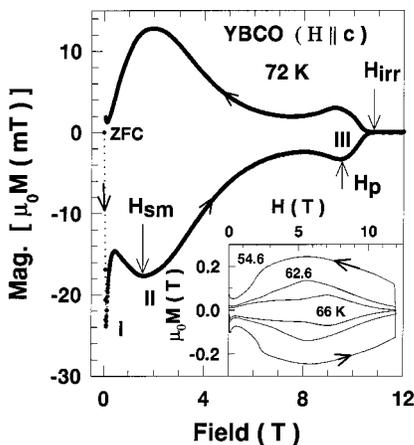


FIG. 2. M - H loop in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ at 72 K. The inset shows the region of anomalous modulation in the M - H loops at 54.6, 62, and 66 K, respectively.

CaRhSn and YBCO at 1.7 and 72 K, respectively. Three peaks (I, II, and III) can be distinctly observed in each sample. In these data, the second (II) and the third (III) peaks could be ascribed to the anomalous variations in J_c . The approach to the first peak (I) just reflects the setting up of the shielding currents $J_c(B)$ within the body of the superconductor as the applied field penetrates deeper into the zero field cooled (ZFC) state. The sharpness of the first peak indicates the rapid decline in $J_c(H)$, once the shielding currents flow in the entire sample. The field at which the forward envelope loop (i.e., the M - H curve from $-H_{\text{max}}$ to $+H_{\text{max}}$ in Fig. 1) meets the virgin ZFC curve (dotted curve) provides a measure of the threshold field H at which the vortices first permeate the entire sample. For instance, in CaRhSn , this value is 17 mT at 1.7 K.

The peaks II and III in CaRhSn and in YBCO point towards the generality of the underlying process. It is natural to identify the latter maximum (III), located at the edge of the irreversibility line, with the notion of the usual PE. The modulation II, therefore, becomes the choice for the SMP. Note that in CaRhSn , $H_{\text{c}2}$ immediately follows H_{irr} (see Fig. 1), whereas in YBCO , H_{irr} lies far below $H_{\text{c}2}$ (the latter is not marked at 72 K in Fig. 2, as it is expected to be > 20 T). The much wider reversible region in YBCO reflects the role of larger thermal fluctuations in high- T_c cuprates.

The relative heights of maxima II and III in Figs. 1 and 2 appear different, however, this does not hide the similarity in the possible transformations of the vortex matter in these two systems in the limited temperature windows. The interaction between vortices, the elastic moduli of vortex solid, the pinning effects, etc., all vary with the field and the interplay between elastic, pinning, and thermal energies^{19,29-31} could favor the stabilization of the vortex phases having different characteristics. The log-log plot of $J_c(H)/J_c(5 \text{ mT})$ vs H in CaRhSn and YBCO at 1.7 and 72 K, respectively. The two curves are vertically separated for clarity. H_{sm} and H_{p} mark the maxima of the SMP and the PE (see Figs. 1 and 2). The graphical similarity in the two curves is striking. We first draw attention to the conspicuous power law regime (PL I), which precedes the onset of the SMP in CaRhSn . A crossover from field independent (or weakly field dependent) to field dependent (notional) power law behavior in $J_c(H)$ is often invoked^{32,33} in weakly pinned samples of low- T_c superconductors to proclaim the arrival of collectively pinned elastic vortex solid phase. If we identify PL I in Fig. 3 with a collectively-pinned ordered solid, then at its lower field end, the crossover to the nearly field independent behavior at $H < 20$ mT could be termed as the approach to the small bundle or individual pinning regime for the dilute vortex array (for $H < 10$ mT, the a_0 of FLL $> 0.5 \mu\text{m}$). Near the upper field end of PL I, the slow decay in $J_c(H)$ could be viewed as a precursor to the effective enhancement in the role of quenched random pins vis a vis interaction between vortices. Following Kokkalis *et al.*,³⁴ we have searched for the memory effects in $J_c(H)$ across the SMP and the PE via a comparison of the tracings of the neighboring minor hysteresis curves^{35,36} and found that the fingerprint of the

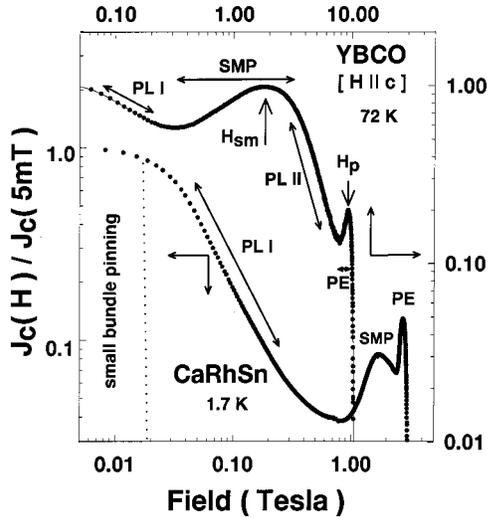


FIG. 3. Log-log plot of $J_c(H)/J_c(5 \text{ mT})$ vs H in $\text{Ca}_3\text{Rh}_4\text{Sn}_{13}$ and $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ crystals at 1.7 and 72 K, respectively. The regions of the small bundle pinning, the collective pinned power law (PL), the second magnetization peak (SMP) and the peak effect (PE) have been identified.

plastic deformation in FLL in CaRhSn and YBCO surfaces near the onset of the SMP (H_{sm}^{on}) and it eventually ceases at H_p , the peak position of the PE. This motivates us to ascribe the SMP as a transformation from an elastic to a plastic regime due to the possible permeation of topological defects, such as the dislocations in an ordered FLL. In the Larkin-Ovchinnikov¹ collective-pinning description, the enhancement in $J_c(H)$ would amount to a progressive shrinkage of the volume of the Larkin domain within which the vortex medium remains elastically pinned. Above H_{sm} , $J_c(H)$ once again decays with H and another power law regime could ensue (see PL I and PL II in the curve for YBCO in Fig. 3). It is therefore reasonable to assert that between H_{sm} and the onset of the PE (H_p^{on}), the balance between interactions and pinning shifts towards the interactions as the vortex density increases further. At the onset of the PE, a marked increase in the memory effects in $J_c(H)$ is witnessed via a characteristic anomaly in the minor hysteresis curves³⁵ and this could be due to the ease with which additional plastic deformations can be caused at the incipient FLL softening stage (i.e., the PE).

To substantiate the above stated assertion further, we show in Fig. 4(a) the field cooled (FC) minor hysteresis curves along with the envelope hysteresis loop in CaRhSn sample at 1.7 K. Note first that the FC minor curves initiated from the FC magnetization values [$M_{FC}(H_{FC})$] overshoot across the envelope loop as the external field is either increased or decreased. The overshooting feature^{37,38} elucidates the inequality, $J_c^{FC}(H) \geq J_c^{ZFC}(H)$ for $H_{sm}^{on} \leq H \leq H_p$, where $J_c^{ZFC}(H)$ corresponds to the current density values along the envelope loop. This inequality implies that a FC vortex state having higher $J_c(H)$ is relatively less ordered³⁷ and has smaller Larkin volume V_c than the corresponding ZFC state. The difference between the saturated (i.e., the peak value) of a FC minor curve (M_{FC}^{sat}) and the correspond-

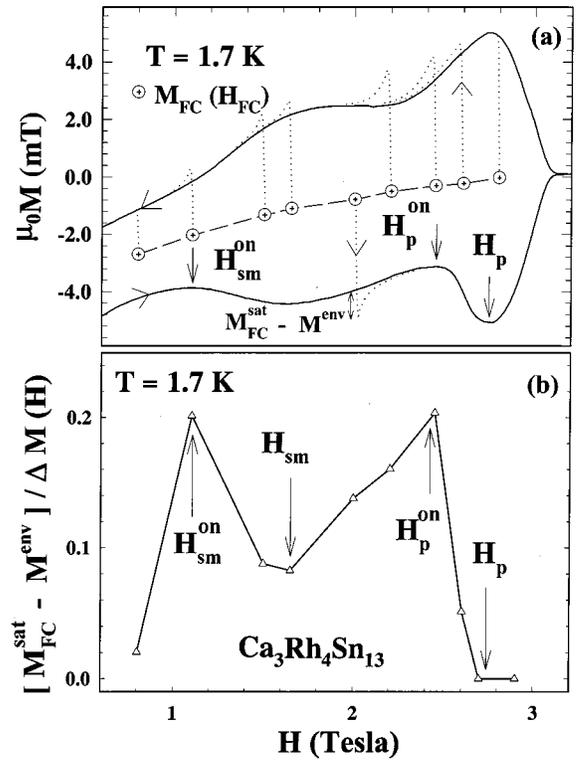


FIG. 4. The panel (a) shows the minor hysteresis curves initiated from different field cooled magnetization values $M_{FC}(H_{FC})$ in $\text{Ca}_3\text{Rh}_4\text{Sn}_{13}$ at 1.7 K. The envelope hysteresis loop is shown as a continuous curve. For $H_{FC}=2 \text{ T}$, the difference between the saturated (i.e., peak) magnetization value (M_{FC}^{sat}) of a FC minor curve and the corresponding magnetization value on the envelope loop (M^{env}) is also indicated. The onset and the peak fields of the SMP and the PE are also marked. The panel (b) shows the plot of the parameter $R_{FC} [= M_{FC}^{sat} - M^{env} / \Delta M(H_{FC})]$ vs H_{FC} in $\text{Ca}_3\text{Rh}_4\text{Sn}_{13}$ at 1.7 K.

ing value on the envelope loop [i.e., $M_{FC}^{sat} - M^{env}$, see Fig. 4(a)] reflects the extent by which a disordered FC configuration differs from the corresponding ZFC configuration. It has been known for long³⁹ that in weakly pinned systems, the FC process attempts to freeze in the state of disorder existing at the peak position of the PE anomaly. With a motivation to ascertain the variation in the state of the spatial order of the FLL with field, it is instructive to view in Fig. 4(b) the plot of the parameter R_{FC} vs H_{FC} , which is defined as³⁴ $R_{FC} = [M_{FC}^{sat} - M^{env} / \Delta M(H_{FC})]$, where ΔM is the hysteresis width of the envelope loop at $H = H_{FC}$. The observed behavior of R_{FC} vs H_{FC} reflects the modulation in spatial order of the FLL between H_{sm}^{on} and H_p through H_{sm} and H_p^{on} . Most significantly, the increase in R_{FC} between H_{sm} and H_p^{on} elucidates that the spatial correlation in FLL enhances in this field interval. The near equality of the peak R_{FC} values at H_{sm} and H_p^{on} further implies that the quality of the spatial order in FLL before the commencement of the two anomalous variations (i.e., the SMP and the PE) in $J_c(H)$ is similar.

Our earlier study³⁸ of the PE in CaRhSn had revealed that an ordered FLL experiences a sudden shrinkage in its corre-

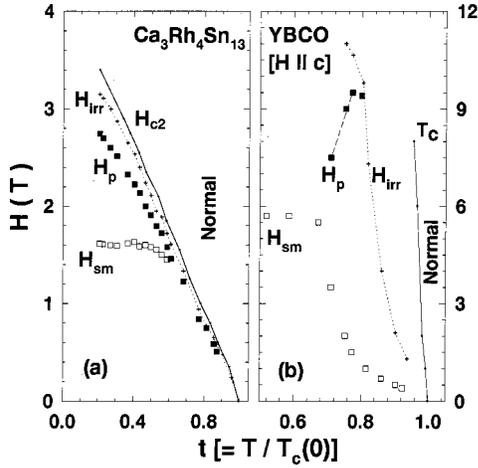


FIG. 5. H_{sm} , H_p , and H_{irr} vs reduced temperature $t [=T/T_c(0)]$ in (a) $\text{Ca}_3\text{Rh}_4\text{Sn}_{13}$ and (b) $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$. The dashed line connecting H_p data points in (b) is just a guide to the eye.

lation volume at the onset temperature of the PE in an isofield scan. The shrinkage in the state of the spatial order in the vortex matter via cumulative plastic deformations presumably reaches its amorphous limit at the peak position (H_p) of the PE, above which all the path dependence in $J_c(H)$ disappears.^{37–40} The pinned amorphous state at H_p is the analogue of the pinned liquid state,²⁸ and the amorphous state loses its pinning ability in a rapid fashion while approaching the irreversibility line.

We may comment on the values of the exponent n in the power law regions I and II in the two materials. In low T_c CaRhSn , n has values of about 1.1 and 1.3 in the regions I and II, respectively. These values compare favorably with those reported in widely studied weakly pinned crystals of 2H-NbSe_2 , where they range between 1 and 1.5.^{21,32} In 2H-NbSe_2 , the pinning is considered to be largely governed by point defects.⁹ In YBCO, the SMP and the PE are observed in the temperature range of 70 K. In this region, giant flux creep is operative. In such a region, J_c is expected to vary as H^{-3} in the large bundle pinning regime (see Fig. 17 of Ref. 28). In our crystal of YBCO, $n \sim -3$ in the region preceding the arrival of the PE.

The magnetization $M(H)$ represents the composite response of the entire sample at a given H (and at a given instance), while the local macroscopic field B exhibits the spatial variation within the sample.⁴¹ Thus, the locus of (H, T) values pertaining to a given characteristic feature in a $M-H$ loop may have only a limited meaning regarding the precise location of the underlying phase transformation (or transition) in the sample.^{41,42} In spite of this caveat, it is instructive to gain information on the T dependence of the two anomalies described above. The insets in Figs. 1 and 2 show how the two anomalies coalesce with the change in T in $\text{CaRhSn}/\text{YBCO}$. Figures 5(a) and 5(b) show the plots of H_{sm} , H_p , and H_{irr} as a function of reduced temperature [$t = T/T_c(0)$] over limited spans in the two systems. In

CaRhSn , $H_{sm}(t)$ appears to be independent of T ($H_{sm} \approx 1.6$ T), while $H_p(t)$ follows the H_{irr}/H_{c2} curves. At about 4.5 K ($t \approx 0.55$), H_p approaches H_{sm} and above 5 K, only the PE survives at $H < 1.6$ T. On the other hand in YBCO, the PE and the SMP broaden and their centers of gravity shift towards each other as T is decreased below 72 K ($t \approx 0.77$). At $T \approx 62$ K ($t \approx 0.67$), the two anomalies merge and only a composite SMP-like feature remains visible (see the inset in Fig. 2). Further reduction in T (< 60 K) flattens out the hump of the SMP, with little movement in its center of gravity. On the other hand, above 72 K ($t \approx 0.77$), the PE peak diminishes rapidly such that only the SMP remains visible above 75 K ($t \approx 0.8$). Also, note that $H_{sm}(t)$ continues to decrease as t increases from 0.65 to 0.93 (i.e., T from 60 to 86 K). This in turn could imply that the regions PL I and PL II (in Fig. 3) could shrink/expand and may even eventually merge as t increases further (i.e., when the SMP becomes dormant). Finally, it may be pertinent to state that in the theoretical framework of Giamarchi and Le Doussal,¹⁹ the phase boundary separating the ordered Bragg glass phase from the dislocation mediated vortex glass phase is field/disorder driven and is insensitive to the temperature variation as is the case for the $H_{sm}(t)$ line [or even $H_{sm}^{\text{on}}(T)$] in CaRhSn . In YBCO, the higher temperature region (> 60 K) in which the thermal fluctuations can renormalize the disordering effects of pins,^{29,30} $H_{sm}(t)$ decreases as t increases, whereas $H_p(t)$ increases. Giller *et al.*⁵ and Nishizaki *et al.*⁶ have surmised that the $H_p(t)$ variation with the positive temperature gradient could be ascribed to the disordering line given by Ertas and Nelson²⁹ and Kierfield *et al.*,³⁰ thereby reflecting the smoothening of the quenched disorder by thermal fluctuations. A theoretical description of the two disordering lines with opposite slopes in the temperature region 60–70 K is awaited.

To conclude, the results in weakly pinned crystals of $\text{Ca}_3\text{Rh}_4\text{Sn}_{13}$ and $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ show that if we identify the regimes of anomalous modulations in $J_c(H)$ with the degree of disorder in the vortex matter and the regime of usual decay in $J_c(H)$ having a notional power law dependence with the collective pinned ordered state, then the features of the coexistence of the SMP and the PE in isothermal scans exemplify a modulation in the degree of plastic deformation in the elastic vortex solid as field increases. A plausible scenario to account it in terms of the competition between the elastic, pinning and thermal energies has been sketched. This scenario finds an echo in the recent observations of Avraham *et al.*,⁴³ who have projected the occurrence of two first order transitions and the presence of an ordered vortex lattice phase sandwiched between disordered phases in an isofield scan in a crystal of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ for $H \parallel c$ via local magnetization measurements.

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- *Corresponding authors. Email addresses: ramky@tifr.res.in / grover@tifr.res.in
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