Vortex matter and its phase transitions

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The mixed state of type II superconductors has magnetic flux penetrating the sample in the form of vortices, with each vortex carrying an identical quantum of flux. These vortices generally form a triangular lattice under weak mutually repulsive forces; the lattice spacing can be easily varied over many orders of magnitude by varying the external magnetic field. The elastic moduli of this lattice are small and this soft vortex matter can undergo phase transitions like normal matter, but with thermal fluctuations and underlying defects playing an important role. We discuss experimental studies on vortex matter phase transitions, with some emphasis on DC magnetization measurements investigating the nature of the phase transition.

ALL the technologically important superconductors, including the so-called high-T_C superconductors (HTSC), belong to the category of type II superconductors. The magnetic field (H) vs temperature (T) phase diagram of these materials comprises two distinct superconducting regions. As depicted in Figure 1, the upper critical field $H_{C2}(T)$ provides the limiting boundary above which superconductivity does not exist. Below the lower critical field $H_{C1}(T)$ we have the perfectly diamagnetic Meissner–Oschenfeld phase. In the region within H_{C1} and H_{C2}, magnetic flux penetrates the superconductor in the form of vortices with each vortex carrying an identical quantum of flux. Traditionally, these vortices are taken to have no structure along their long axis parallel to the applied H. The superconducting order parameter has a minimum at the centre of each vortex, and the field has a maximum, as depicted in Figure 2. The field decays as one moves away from the vortex centre; accordingly the vortices carry identical cylindrical current shells at their periphery. These vortices repel one another as expected by the identical current shells and Abrikosov had argued that they form a triangular lattice. The lattice spacing decreases continuously as the applied field is raised from H_{C1} to H_{C2}. The dissipationless current-carrying applications of superconductors require an understanding of vortex matter and this has been continuously receiving attention[1]. In this paper we focus, however, on some studies regarding phase transitions amongst different phases of vortex-matter. The density of vortex matter can easily be varied over orders of magnitude, and vortex–vortex interactions are weak. This makes vortex matter, *a la* colloids[2], a potential testing ground for our understanding of structural phase transitions.

Brief history of early studies

Questions regarding structural changes in vortex matter were first addressed over three decades ago, soon after Abrikosov's prediction of a triangular vortex lattice. We shall briefly recapitulate those results as relevant today.

Assuming the vortices to have no structure along their long axis, the triangular vortex lattice will have only three stiffness moduli. These elastic constants become weak as H approaches H_{C2} because the modulation in local field, which dictates the electromagnetic interaction between vortices, falls towards zero. The interactions also tend to zero close to H_{C1} because the vortex separation rises. The elastic constants of vortex matter are then many orders of magnitude smaller than those of normal matter[3], and thermal fluctuations as well as defects in the underlying superconducting solid become important in the free energy of the vortex solid. This can result in the vortex matter developing a fluid-like structure close to H_{C1}[4], as well as close to H_{C2}[5]. Labusch[6] had shown that the shear modulus goes parabolically to zero as the applied field nears H_{C2}, and

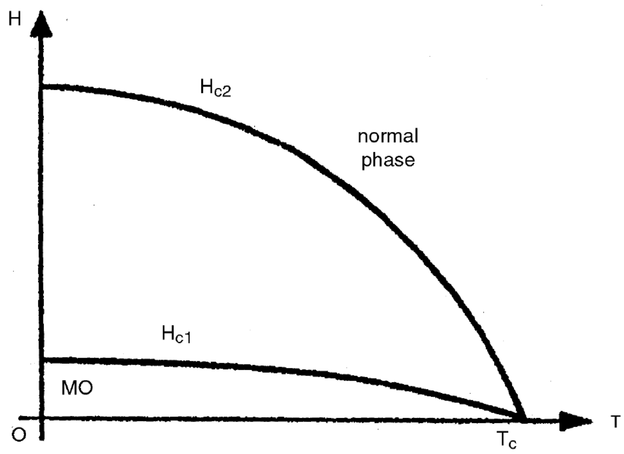

Figure 1. The superconducting phase exists below the $H_{C2}(T)$ line. The perfectly diamagnetic Meissner–Oshenfeld phase exists below the $H_{C1}(T)$ line. The region between the $H_{C1}(T)$ and $H_{C2}(T)$ lines is the mixed state where vortex matter exists.

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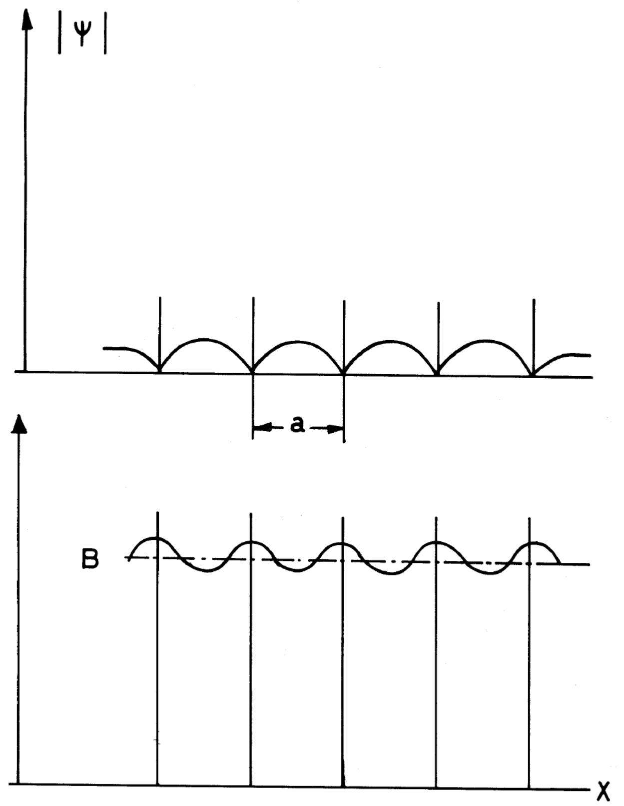

Figure 2. The order parameter $|\psi|$ and the local induction B are plotted as a function of position in space. The vortices are a distance a apart, and their centres (location is indicated by the vertical lines) correspond to peaks in B of magnitude H_{C2} and to the order parameter dropping to zero. The amplitude of the oscillation in B falls as H approaches H_{C2}.

had concluded that 'This would indicate a tendency of the fluxoid assembly to develop a fluid-like character near H_{C2} instead of forming a regular lattice'. Labusch also recognized[6] the role of crystal imperfections (in the underlying superconducting solid) in deciding whether or not the vortex matter would actually transform to a fluid-like state. Eilenberger[7] had earlier studied thermodynamic fluctuations of the order parameter for H near H_{C2} and had concluded that 'the vortex lattice melts at a certain external field strength below H_{C2}. This would be rather interesting if it were observable experimentally'. His prophecy proved right after nearly thirty years!

For very low vortex density near H_{C1}, the shear modulus again becomes small and a fluid-like response was predicted[4]. Trauble and Essmann, who had pioneered the so-called decoration technique for imaging vortex structures, have at least twice reported such interesting observations. In their studies on Pb-(6 at%) In, Trauble and Essmann[8] found that above a critical vortex lattice spacing of d_C = 8000 Å (corresponding to a magnetic induction of about 4 mTesla) 'the flux-line pattern resembles an instantaneous pattern of a two-dimensional fluid'. Similarly, in Pb-Tl, Essmann and Trauble[9] observed a fluid-like structure for magnetic induction below 2.5 mTesla.

These early works on vortex matter transforming from a solid to a fluid near both H_{C1} and H_{C2} are, for reasons that are unclear, not much discussed in current literature. This is not true of predictions made around the same time by Fulde and Ferrel[10], and by Larkin and Ovchinnikov[11], that in paramagnetic superconductors the superconducting order parameter would develop modulations along the length of vortices. This would occur at fields close to H_{C2} and was predicted[12] to be a first order phase transition (FOPT) to the Fulde–Ferrel–Larkin–Ovchinnikov (FFLO) state. The FFLO theory is still used by theorists studying coexisting superconductivity and weak magnetism[13]. The prediction of a FOPT has motivated experimentalists in recent years, as will be discussed later.

Theoretical background

Possible phase transitions

Many developments of concepts in phase transitions have also influenced recent studies on vortex matter. Dynamic phase transitions have been proposed in driven lattices[14] with velocity being inversely related to an effective temperature. Experimental studies have shown very interesting results on driven vortex matter and its effective phases (see refs 15 and 16 and references therein). This new and interesting field is outside the scope of the present paper.

We now briefly enumerate recent theoretical ideas on possible phase transitions in stationary vortex matter. It has been argued (see Blatter[17] and references therein) that a pure type II superconductor must show a reentrant liquid phase as depicted in Figure 3. The triangular

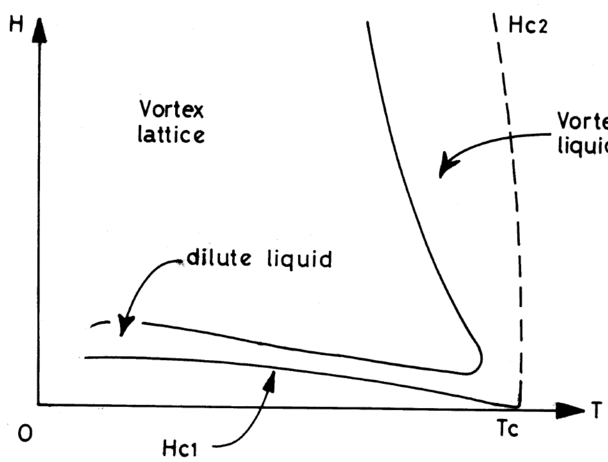

Figure 3. Theoretically proposed phase diagram showing reentrant melting, with vortex solid encompassed by the vortex liquid.

vortex lattice is expected to melt with rising T via a FOPT. It would also melt both as H is raised and as H is lowered. Sophisticated theories have also argued that in the presence of weak quenched disorder, the low-H low-T phase would be a Bragg glass (BG) where the position correlation function decays as a power law. This BG phase of vortex matter would also melt with increasing temperature via a FOPT. With increasing field there would be a sudden proliferation of dislocation lines and the BG phase would transform to a vortex–glass (VG) phase. There has been a recent theoretical prediction[18] that this BG to VG transition would also be a FOPT. We shall discuss experiments investigating the thermodynamic nature of these transitions in the next section.

There has also been a lot of work on periodic vortex lattices that are not triangular. Kogan and collaborators[19] have argued that non-local effects in the vortex interaction can change the vortex lattice symmetry, to other than triangular, as a function of H and T. Such changes have been observed experimentally in the rare-earth nickel borocarbides[20], in the HTSC material $YBa_2Cu_3O_7$[21], as well as in V_3Si[22]. This is presently another very active area, but we shall not cover it in this paper because our emphasis is on experimental studies concerning the thermodynamic nature of vortex matter phase transitions.

On signatures of first order phase transitions

We first recall that a phase transition differs from the colloquial 'transformation' in that a thermodynamic parameter will change discontinuously when the phase boundary is crossed by varying a control parameter like T or H. To report a FOPT along a $T_C(H)$ line, one needs to observe a discontinuous change in entropy (i.e. observe a latent heat L), or in magnetization (i.e. in vortex volume), as one crosses the $T_C(H)$ line by varying either of the control variables T or H. The FOPT is firmly established if the magnetization jump ΔM and L satisfy the Clausius–Clapeyron relation; for this we also need $\mathrm{d}T_C/\mathrm{d}H$, and thus need to locate $T_C(H)$ over a finite region of H–T space. If we have only observed a discontinuous change in some thermodynamic variable that is a second or higher derivative of free energy (like susceptibility), then we can report a (continuous) phase transition only. If we have observed only two regions of H–T space where the phases have widely differing thermodynamic properties, then more rigorous experimental checks should be sought before we can even assert that the phase transformation occurs through a phase transition.

It is, however, common (and justified) to report less rigorous signatures as possible indicators of a FOPT. Amongst these are coexistence of two phases, and supercooling/superheating across the $T_C(H)$ line. The latter feature presents itself as hysteresis when locating the phase boundary (as a function of H or T) via a sharp change in the measured property. To understand the need for cross-checks after observing these indicators, we consider a scenario[23] in which hysteresis may be seen without a FOPT. Let us say that we are pressure cycling a material which is very viscous (or is an amorphous solid), and there is a pressure range where the volume changes very strongly with pressure, but not discontinuously. If we are changing pressure fast then the observed volume is not the equilibrium volume, but lags behind because of the high viscosity. A similar effect is expected when pressure is being reduced. Thus we would observe a hysteresis loop that is purely kinetic in origin. Kinetic hysteresis can be seen when molecules in amorphous solids (or vortices in hard superconductors) exhibit hindered kinetics, and equilibrium can be reached only over times much longer than experimental time scales. This is very relevant for vortex matter in hard superconductors where M–H hysteresis is understood using Bean's critical state model; which has hindered kinetics of vortices (or pinning of vortices) as its cornerstone. Metastability due to slow kinetics may also be possible at a second order phase transition due to critical slowing down. Hysteresis and metastability, by themselves, are thus arguable signatures of a FOPT.

The $T_C(H)$ line can be crossed by varying either of T or H, and hysteresis in locating the FOPT can be seen with either control variable. We have argued recently[24] that for a FOPT the range of supercooling (or extent of hysteresis) is more when T is varied at constant H, than when H is varied at constant T. This thus provides a necessary condition for hysteresis to be attributed to a FOPT. A series of other predictions have been made regarding metastabilities across a FOPT under different experimental protocols[24,25]. One that is relevant here is that repeated cycling of magnetic field introduces a large fluctuation energy that depends linearly on the number of cycles, and nonlinearly on the amplitude of the cycling field. These two parameters are also, usually, not strictly controlled in AC measurements. Further kinetic hysteresis will be (more) dominant in (higher frequency) AC measurements. We shall, in this paper, discuss mainly DC measurements where both these complications are minimal.

Overview of experimental results

Vortex lattice melting

As Eilenberger[7] had prophesied, vortex lattice melting turned out to interest many researchers. In $YBa_2Cu_3O_7$, all features of a FOPT, viz. hysteresis, vortex–volume discontinuity, and latent heat have been observed and the Clausius–Clapeyron relation has also been established. We summarize below a sequence of key observations (in our opinion) that established this FOPT.

The resistivity of $YBa_2Cu_3O_7$ measured in a magnetic field showed a sharp step while dropping to zero, and the resistivity step was accompanied by hysteresis both with T and with H as control variables[26–28]. This hysteresis was interpreted as a signature of a FOPT, and ascribed to vortex lattice melting. Welp *et al.*[29] stressed that transport measurements do not probe the defining characteristics of a magnetic FOPT, and they showed that the resistivity step was accompanied by a step in the magnetization, with the height of the step rising as the transition point moved to lower T or higher H. The jump in magnetization they observed, however, occurred over a field range of the order of 0.1 Tesla. This width has been understood as an effect of sample geometry in these bulk measurements of magnetization; Zeldov *et al.*[30] showed in their studies on $Bi_2Sr_2CaCu_2O_8$ that the width becomes negligible when local measurements are made using microhall probes. Soon thereafter, Schilling *et al.*[31] measured the latent heat across the transition in $YBa_2Cu_3O_7$, in conjunction with measuring the jump in magnetization. Both these measurements were made over a wide region of the melting line, and they showed that over this entire region the Clausius–Clapeyron relation was valid. Vortex lattice melting was thus firmly established as a FOPT.

True to Eilenberger's prophecy, vortex lattice melting has since been investigated in many superconductors and remains a very active area. Observation of coexisting liquid and solid vortex matter has also been reported recently[32]. We should mention here that the phase diagram depicted in Figure 3, with reentrant melting, has also provoked experimentalists. A reentrance of the peak-effect (in J_C vs H), that resembles the reentrance of the theoretically proposed melting curve, has been observed in a few samples of 2H-$NbSe_2$[33,34]. Questions regarding the influence of point defects, as well as of extended line defects, on melting of vortex matter also remain and the results will have some relevance to the behaviour of normal matter (see e.g. refs 35 and 36).

First order solid to solid phase transition

The role of topological defects in producing an amorphous vortex solid (as H is raised towards H_{C2}) has been discussed early in conventional superconductors[37], and an amorphous solid has been observed recently by neutron scattering[38]. These studies have not, however, addressed the nature of the phase transition (if any) between the solid phases. While there are many reports of such a transition in the HTSC compounds[39–41], both theory and experiments have begun questioning the nature of the transition (i.e. whether or not it is a FOPT) only very recently. Experimentalists motivated by the FFLO theory, which predicts a solid-to-solid FOPT, were testing for the nature of the phase transition earlier, and we shall first describe these experiments.

In a generalized version of the original FFLO theory, it was argued that the vortices are segmented into short strings with concomitant enhancement of pinning[42,43]. This results in a peak-effect (PE) in J_C vs H, or in the M-H hysteresis curve, as H is raised towards H_{C2}. $CeRu_2$ was a candidate material for an FFLO state ever since the discovery of a magnetic anomaly near H_{C2}[44]. The first thermodynamic signature indicating that the onset of PE is a FOPT came through the observation that the PE starts at a field H_a^* on increasing field, but disappears at a lower field H_d^* on decreasing field[45–47]. The different values in the field-increasing and field-decreasing cases was taken as a signature of the hysteresis expected in a FOPT. We have attempted to identify other measurable signals of a FOPT, and the FFLO theory motivated us, possibly as a red herring. We shall now describe our DC magnetization studies[48–50] on $CeRu_2$, and shall mention similar studies by other groups on $CeRu_2$[51,52], $NbSe_2$[52–55] and $YBa_2Cu_3O_7$[55–57]. The motivating theories for the studies on the last two materials were not FFLO but vortex lattice softening and BG to VG transition.

There has been no reported observation so far of a latent heat associated with the onset of PE in any of these materials. A step in the vortex volume has been reported by us at the onset of PE in $CeRu_2$[49], and by Ravikumar *et al.*[53] in $NbSe_2$. But these measurements are tedious compared to similar measurements for vortex lattice melting, because the equilibrium magnetization has to be extracted[58] from experimental M–H curves that are hysteretic. Since hysteresis in locating the onset of PE had been reported as a signature of a FOPT[45–47], it was natural to look for supercooled states; and these were reported soon thereafter[48,49]. Conventionally, supercooling of a liquid is established by measuring diffusivity which would decrease by orders of magnitude if the solid is formed, but would fall smoothly as the liquid was supercooled below the melting point. Since the FFLO state is characterized by higher pinning or higher J_C, supercooling can be confirmed by measuring minor hysteresis loops[48–50] that are related to J_C through the critical state model[59]. [This technique of measuring minor hysteresis loops (MHLs) has the added advantage that one can ensure that hysteresis is due to bulk pinning[60] (see Figure 4). One can also ensure that no artefacts have been introduced by the slight inhomogeneity of the magnetic field[47].] The two phases on either side of the PE onset have different J_C values, and a supercooled metastable phase at a (H, T) point will produce an MHL that is distinct from the

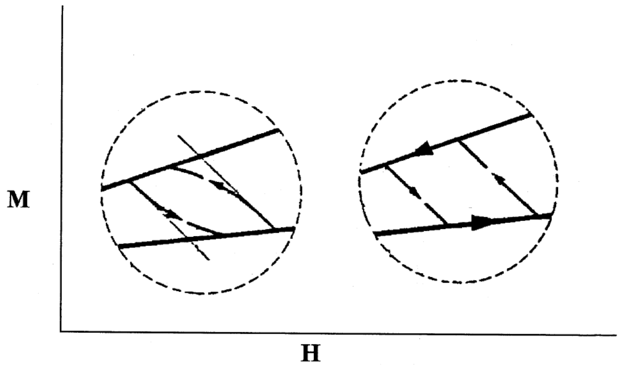

Figure 4. The behaviour of minor hysteresis loops drawn between the envelope curves is indicated by thicker lines. As indicated in the right circle, the minor loops are straight lines if hysteresis is due to surface currents. The left circle displays minor loops when hysteresis is due to bulk pinning, as in critical state model. The minor loops are then continuously nonlinear, breaking away from the straight line as soon as they leave the envelope curves.

MHL produced by the equilibrium phase. The MHLs will thus be history-dependent in the region of (H, T) space where supercooled metastable states can be made to exist. This feature of history-dependent MHLs was observed by us in various polycrystalline samples of $CeRu_2$ and was invoked by us as evidence of supercooling and hence of a FOPT[48–50]. History-dependent MHLs have subsequently been observed in single crystal samples of $CeRu_2$[51,52,61], and also in $NbSe_2$[52–55] and $YBa_2Cu_3O_7$[55–57] single crystals. We must mention here that not all samples of $YBa_2Cu_3O_7$ show history-dependant MHLs[56].

We were able to track this FOPT line in $CeRu_2$ over a factor of four in vortex density, and the various histories under which MHLs were observed corresponded to crossing this FOPT line through various paths in (H, T) space. It was clear that supercooling persisted farther when T is lowered at constant H, than when H is lowered in constant T[60]. This observation is consistent with the theory for supercooling described in the previous section. This theory makes many other predictions, one of which we now highlight. If $H^*_{FC}(T)$ is the limit to which supercooling can persist when T is lowered in constant H, and $H^*_d(T)$ is the limit when H is lowered at constant T, then it was predicted[24,25] that since T_C falls with rising H, the region separating $H^*_{FC}(T)$ and H^*_d (T) will broaden as H rises (or as T falls). This prediction is depicted schematically in Figure 5 *a*. We measured[62] these limits of supercooling in a single crystal sample of $CeRu_2$, and the results shown in Figure 5 *b* are consistent with predictions.

As we had argued earlier, hysteresis can be kinetic in origin and observation of hysteresis (or of phases coexisting over finite time scales) may not be taken as a sufficient indication of a FOPT. Naïve arguments[25] suggest that if hysteresis is kinetic in origin then the

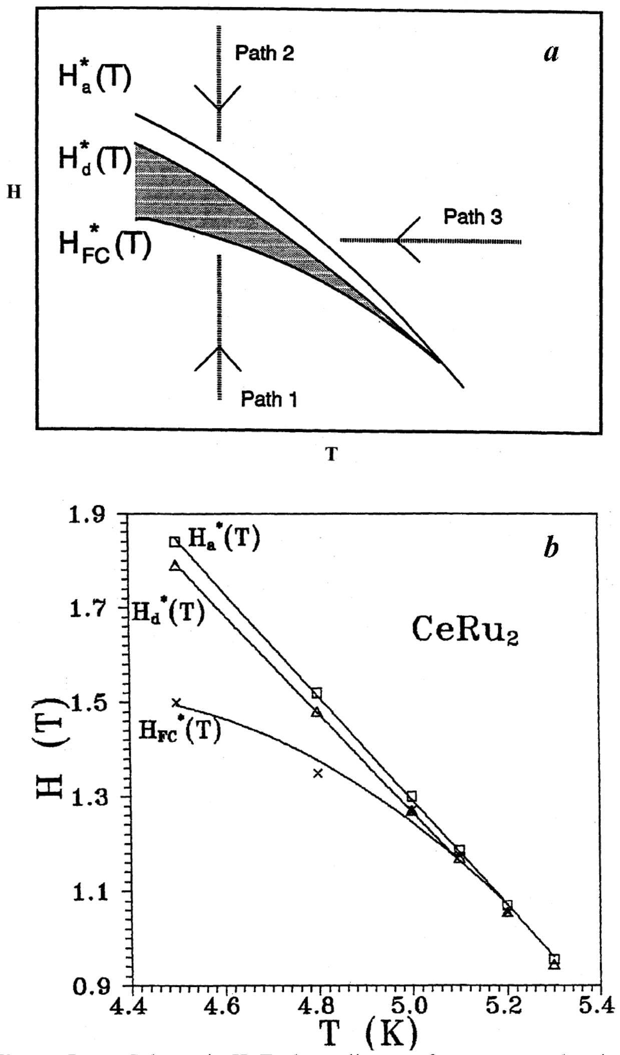

Figure 5. ***a***, Schematic H–T phase diagram for a system showing a FOPT. Following ref. 24, path 1 (increasing H at constant T) leads to the line H_a*. Path 2 (decreasing H at constant T) leads to the line H_d*, while path 3 (decreasing T in constant H) leads to the line H^*_{FC}. ***b***, The experimentally observed H–T phase diagram in the case of $CeRu_2$ (ref. 62). Lines through the data are drawn as guide-to-the-eye.

path dependence (in H–T space) of the accompanying metastability would have an inequality of sign opposite to that predicted for a FOPT. This assumes some significance in the context of experiments to be discussed below.

We finally draw attention to two recent experiments that conclude that the BG to VG transition in $Bi_2Sr_2CaCu_2O_8$ is a FOPT. Gaifullin *et al.*[63] report a sharp change in the plasma frequency at the BG-VG boundary and, since the change here is at least as sharp as that across the melting transition, they conclude that BG to VG is a FOPT. Using magneto-optic studies, van der Beek *et al.*[64] and Giller *et al.*[65] observed coexistence of the BG and VG phases, as well as supercooling of the VG phase. Van der Beek *et al.* also conclude that the BG to VG is a FOPT. We must point out that they report coexistence for a few seconds after a sudden lowering of the applied field, and this is *not* seen to persist beyond about ten seconds. Going back to our discussion on hindered kinetics (and kinetic hysteresis) in the previous section, question arises whether van der Beek *et al.* are reporting a slow transient phenomenon. We then also recognize that the measurements of Gaifullin *et al.* are at high frequency. In the view of the present authors, steady state measurements are still necessary to establish the nature of the BG to VG transition. We should mention here that we have earlier failed to observe history-dependent MHLs in $Bi_2Sr_2CaCu_2O_8$ but ours were DC measurements with a waiting time of about 100 seconds. It is also possible that the supercooled state in $Bi_2Sr_2CaCu_2O_8$ is very fragile and fluctuations induced during the measurement process shattered the supercooled VG phase. The nature of the BG to VG transition clearly needs more experiments, specially looking for the extent of metastability after following different paths in (H, T) space.

Summary and conclusions

We have discussed the current status of DC magnetization studies on phase transitions in stationary vortex matter. Vortex lattice melting is established as a FOPT, while detailed experimental studies show supercooling across a solid-to-solid transition of vortex matter, indicating another FOPT. We have predicted a specific path-dependence in the supercooling possible when T and H (or vortex density) are varied across a FOPT, and the data on $CeRu_2$ are consistent with this. While our predictions are valid for normal matter like the water-to-ice transition, vortex matter phase transitions are seen over much larger variations in density and provide experimentally easier testing grounds.

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