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Possibility of Kauzmann points in the vortex matter phase diagram of single crystal $YBa_2Cu_3O_{7-\delta}$

S.B. Roy [a,b,*], Y. Radzyner [a], D. Giller [a], Y. Wolfus [a], A. Shaulov [a], P. Chaddah [b], Y. Yeshurun [a]

[a] Department of Physics, Institute of Superconductivity, Bar-Ilan University, 52900 Ramat-Gan, Israel
[b] Low Temperature Physics Laboratory, Centre for Advanced Technology, Indore 452013, India

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Abstract

We highlight interesting thermomagnetic history effects across the transition line between the quasi-ordered and disordered vortex states in single crystal $YBa_2Cu_3O_{7-\delta}$ (YBCO), and argue that these features are indicative of the first-order nature of the transition line. We suggest that the destruction of the ordered vortex state in YBCO leading to vortex liquid (at high temperatures and low fields) and amorphous vortex solid (at low temperatures and high fields), takes place along a unified first-order transition line. The non-monotonic behavior of this first-order transition line gives rise to the possibility of more than one Kauzmann point where the entropies of the ordered and disordered vortex states are equal. In the high temperature region, one may order the vortex lattice by warming it, giving rise to an inverse melting effect.

Keywords: First-order transition; Ordered and disordered vortex solid; Vortex cores; Kauzmann point; Entropy crisis

1. Introduction

In a recent report Avraham et al. [1] have shown that the destruction of the quasi-ordered vortex lattice or Bragg glass [2] in single crystal samples of $Bi_2Sr_2CaCu_2O_8$ (BSCCO) takes place along a unified first-order transition line. Two different types of energy—thermal energy and pinning energy—actually compete with the elastic energy leading to the destruction of the ordered vortex lattice. At low temperatures and high fields pinning dominates, leading to a field/disorder induced destruction of the ordered vortex lattice. At high temperatures this unified transition line changes its character from disorder induced transition to thermally induced melting. The apparently unusual finding is the non-monotonic nature of this first-order transition line, leading to the paradoxical situation that in a certain field–temperature (B–T) window the 'ordered' vortex state has larger entropy than the 'disordered' vortex state. This in turn has the interesting implication that a crystal transforms into liquid or amorphous state on decreasing the temperature. Such a

* Corresponding author. Present address: Low Temperature Physics Laboratory, Centre for Advanced Technology, Indore 452013, India. Fax: +91-731-481426.
E-mail address: sbroy@cat.ernet.in (S.B. Roy).

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situation is quite rare but not unknown, one classic example being the melting curve of ^{3}He showing a pressure minimum [3]. Similar situation also exists in spin lattice systems [4] where the spin-glass state transforms into a long-range magnetic ordered state with the increase in temperature. However, such a transition is known to be a continuous transition with definite critical response [4].

The same non-monotonic character of the transition line between two kinds of vortex solids has been reported earlier for $YBa_2Cu_3O_{7-\delta}$ (YBCO) single crystals [5,6]. In addition the presence of metastability has also been highlighted across this phase transition [6,7]. We extend this study to argue that the vortex solid–solid transition line in YBCO is also a first-order transition line, and there exists a situation of 'ordering by heating' in a certain B–T window of YBCO as well. The non-monotonic nature of the phase transition line leads to one minimum and one maximum point where the entropies of the disordered and ordered solids are equal. Such equal entropy points are known as Kauzmann points in the literature (Ref. [8] and references therein). This is related to the concept of 'entropy crisis' in supercooled glass forming liquids where in principle below a temperature known as Kauzmann temperature (T_{Kauzmann}) a disordered metastable liquid would appear to have an entropy lower than that of an ordered solid. This 'entropy crisis' is usually avoided by the formation of a glassy state of unique configuration at a temperature $T_g > T_{\mathrm{Kauzmann}}$.

2. Experimental

An array of 11 10×10 μm^2 Hall sensors (sensitivity better than 0.1 G), consisting of a GaAs/Al–Ga–As two-dimensional electron gas layer was in direct contact with the surface of a $0.5 \times 0.3 \times 0.02$ mm^3 untwinned YBaCuO single crystal ($T_C \approx 93$ K) [9]. The Hall-sensor array allows the study of local vortex behavior with high field sensitivity and high spatial resolution. An additional advantage of this technique is that the actual value of the local induction, B_z, is obtained. In global measurements, only the external applied field is known, while the internal induction is difficult to evaluate owing to complicated demagnetization effects [10]. The results reported in this paper utilise two sensors, one close to the sample's center and another just outside the sample. The difference in the local induction measured by these two sensors is directly related to the magnetization of the sample. (This shall be referred to as 'magnetization (m)' in the rest of this paper.)

We shall use the 'minor hysteresis loop' (MHL) technique [11] to study the first-order transition line. This technique consists of drawing MHL at various points on the envelope M–H curve by reversing the field direction. Away from a vortex matter phase transition line these MHLs follow the predictions of Bean critical state model (CSM). The deviations from this standard behaviour occur near a first-order vortex solid–solid transition and various features of these MHLs can be used to study the phase coexistence and metastability across such transition [11]. We believe that this technique, although less rigorous than the equilibrium thermodynamic measurements like calorimetry for confirmation of a first-order transition, belongs to the class of experiments which can investigate the phase coexistence and supercooling across a first-order transition. (The equilibrium thermodynamic confirmation of a first-order transition becomes difficult if either the latent heat is small [12], or the first-order transition is broadened by disorder [13].) This MHL technique has now been used to study the nature of the vortex solid–solid transition in low temperature superconductors like $CeRu_2$ [14–16], $NbSe_2$ [17,18] and V_3Si [19], in high temperature superconductors such as YBCO [6] and LaSrCuO [20], and most recently in MgB_2 [21].

3. Results

A typical field dependence of magnetization showing peak effect (PE), which we use to track the vortex solid–solid phase transition line from a quasi-ordered vortex solid to disordered vortex solid, is shown in Fig. 1. The relatively broad second peak, measured in untwinned YBCO, exhibits a sharp onset at B_{onset} and a sharp change of slope at B_{kink} [5,22]. These two features occur at

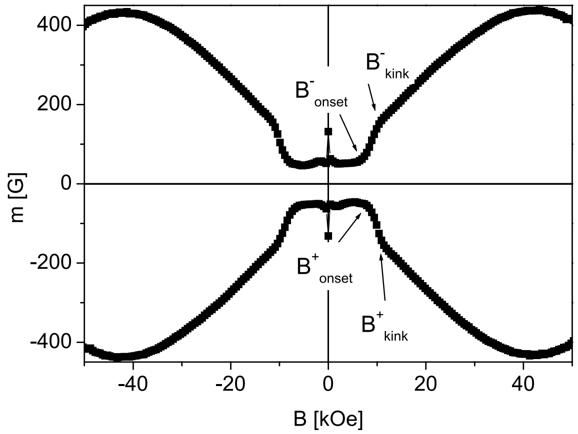

Fig. 1. Magnetization loop with PE for the YBCO crystal at $T = 55$ K. Four characteristic fields B^{+}_{onset}, B^{-}_{onset}, B^{+}_{kink} and B^{-}_{kink} are identified and marked in the figure.

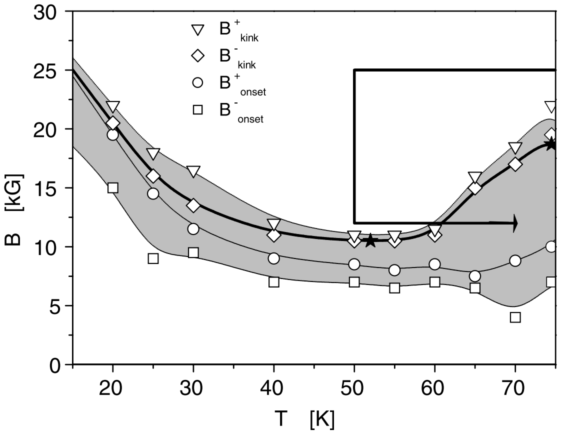

Fig. 2. Magnetic phase diagram for YBCO crystal showing B^{+}_{onset}(circle), B^{-}_{onset}(square), B^{+}_{kink}(triangle), B^{-}_{kink}(diamond) lines. As discussed in Ref. [6] $B^{-}_{\mathrm{kink}}(T)$ line is the vortex solid–solid phase transition line $B_{\mathrm{SS}}(T)$. Also marked are the Kauzmann points (stars). Path marks the second field-cooling experimental protocol (see text for details).

different fields on the ascending and descending branches of the magnetization loop, resulting in four different characteristic fields: B^{+}_{onset}, B^{-}_{onset}, B^{+}_{kink} and B^{-}_{kink}, marked by arrows in Fig. 1. It has been observed that an abrupt change in the external field causes the injection of a transient disordered state into the sample [23]. When the thermodynamic conditions dictate a quasi-ordered state, the injected transient disordered state relaxes into a quasi-ordered state at a rate decreasing to zero as the induction approaches the solid–solid thermodynamic transition field B_{SS}. Since our experimental procedure involve steps of 500 Oe between adjacent measurements, a generation of transient disordered state can be expected after each step. As B^{+}_{onset} is approached the lifetime of the disordered state is comparable to the time window of the measurement and a larger persistent current is measured, indicating the existence of a disordered state. At B^{+}_{kink} the disordered phase becomes the stable thermodynamic phase. B^{-}_{kink} cannot be associated with the lifetime of the transient disordered state. This is because above the metastability region thermodynamics dictate a disordered state, so that the phase introduced by the change of field does not alter the phase already existing in the sample. Thus it is natural to identify B_{SS} with B^{-}_{kink} (Ref. [6]). After crossing $B_{\mathrm{SS}}(T)$ line vortex matter gradually becomes more ordered until B^{-}_{onset} is crossed and the matter becomes completely ordered. Collating this characteristic field B^{-}_{kink} at various temperatures from our isothermal magnetization studies we present a *B–T* phase diagram in Fig. 2 showing this $B^{-}_{\mathrm{kink}}(T)$ line. This is similar to the (*B–T*) phase diagram that has been reported earlier [6], but is reproduced here again to make the present study a self contained one.

It is apparent from Fig. 2 that the non-monotonic nature of the phase transition line is more prominent in comparison to BSCCO [1]. The slope of the transition line changes sign twice as a function of temperature, first at around 50 K going from negative to positive, and then at around 75 K back to negative again. We shall now concentrate on the 50–75 K regime of this phase transition line where, akin to that in BSCCO [1], exists the interesting suggestion of a transition from the disordered solid to ordered solid achieved by heating. We present results in the form of MHLs obtained after preparing the vortex state following two distinct experimental protocols:

(1) Zero field cool (ZFC) the sample to the temperature of measurement and then increase the field to go to the vortex solid–solid phase transition region denoted by the shaded area in Fig. 2.

The field is then lowered towards zero so that an MHL is obtained.

(2) Cross the vortex solid–solid phase transition line by varying temperature in the presence of an external field. To do this in the 50–75 K regime, which had not been explored before, we cool the sample from above T_C to 50 K in the presence of external field of 25 kOe. We then lower the field to the target value at 50 K, and then increase the temperature to the temperature of measurement. This is the counterpart of the step-down procedure used in Ref. [6], where only the low-temperature region was explored.

In Fig. 3 we present MHLs obtained under the protocol 1 at 70 K. At this temperature the kink cannot be easily seen, but is clearly defined by a maximum in dm/dB. These MHLs are obtained by terminating the field increasing cycle of the m–B curve at various points in the field regime $B^-_{onset} \leqslant B \leqslant B^+_{kink}$. Within the Bean CSM (which explains well the irreversible magnetization of type-II superconductors) such MHLs are expected to meet the envelope M–B curve after the field is decreased by an amount $2B^*$, where B^* is the field for full penetration [24] at that particular B. In fact the MHLs drawn at various fields $B > B^+_{kink}$ show this expected behaviour. However, the behaviour changes markedly at the onset of the PE regime,

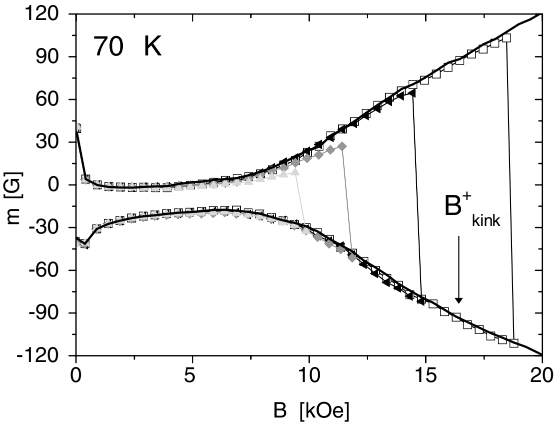

Fig. 3. Magnetization loop (represented by solid line) and ZFC MHLs following the experimental protocol 1 (see text for details) at 70 K. The MHLs were initiated by reversing the field cycling at 10 kOe (up triangles), 12 kOe (diamonds), 15 kOe (down triangles) and 19 kOe (squares).

and the MHLs obtained by procedure 1 (ZFC) saturate without reaching the the upper envelope curve (see Fig. 3). 'The "amount of hysteresis" Δm, i.e. the difference between the magnetization of the complete loop and the MHL at a given field, reflects the changes in local magnetic induction [25]: $\Delta m > 0$ reflects the presence of domains of disordered vortex solid (with enhanced J_C) in the sample, whereas $\Delta m < 0$ implies the presence of domains of ordered vortex. It is clear from our data that the vortex disordered solid (with enhanced J_C) starts nucleating at $B > B^+_{onset}$ and its formation is complete only at B^+_{kink} where the MHLs saturate only on reaching the upper envelope curve. This kind of nucleation and growth of the enhanced pinning state are typical characteristics of a first-order transition, and have earlier been observed in low temperature superconductors like $CeRu_2$ [11,14–16] and $NbSe_2$ [17,18], V_3Si [19], and high temperature superconductors like YBCO [6] and LaSrCuO [20].

In Fig. 4 we present MHLs at 70 K obtained under the experimental protocol 2. The MHLs, obtained both by increasing and decreasing B, overshoot the envelope curve. This in turn implies that flux pinning obtained in this manner is more than that obtained in crossing the vortex solid–solid transition line by isothermal field variation. It has been argued earlier that extent of metastability (i.e. supercooling/superheating) associated with a first order transition is more if the transition line is crossed by the variation of temperature in presence of a constant magnetic field in comparison to the situation where the line is crossed by isothermal variation of the applied field [26]. This is because the variation of the magnetic field produces fluctuation which drives the metastable state in the local minimum of the free energy curve to the stable state across the energy barrier. We argue that the observed overshooting of the MHLs in Fig. 4 is thus another indication of the first-order nature of the vortex solid–solid transition line.

It is to be noted here that the metastability across the first-order vortex solid–solid transition line has been highlighted recently in BSCCO [23,27]. In fact the associated irreversibility in this transition region was removed by using an applied ac field H_{ac} to reveal the step-jump in the

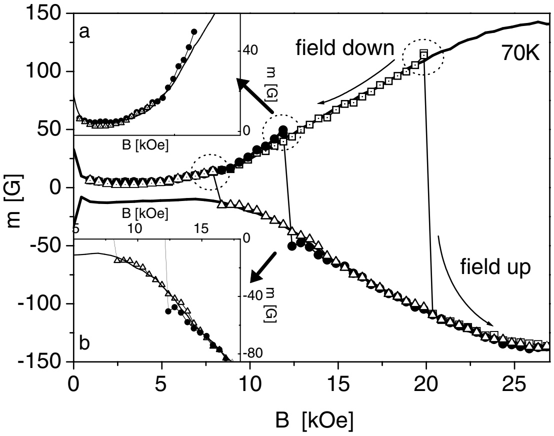

Fig. 4. Magnetization loop (represented by the solid line) and FC MHLs following the experimental protocol 2 at 70 K. The sample is cooled from above T_C to 50 K in the presence of an external field of 25 kOe. The field is then lowered to target fields $H = 8$, 12 and 20 kOe at 50 K and the temperature is raised to the temperature of measurement i.e. 70 K. The MHLs are obtained starting at each of these target fields both by lowering and raising field. Starting points of the MHLs are marked with dotted circle and the directions of the MHLs are shown as field up and field down arrows for the representative target field $H = 20$ kOe. The overshooting of the FC MHLs beyond the complete magnetization loop (solid line) is highlighted in the insets of the figure both while raising (see lower inset) and lowering (see upper inset) the field at 12 kOe.

magnetization, which in turn was used to establish the first-order nature of the transition line [1]. In contrast we use the metastable characteristic across the transition line itself to identify its first-order nature.

4. Discussion

Combining the present study as well as the earlier ones on YBCO [5,6] we argue that as in BSCCO the extended $B_{SS}(T)$ line coincides with the melting line of the vortex ordered solid $B_M(T)$ at high temperatures. Hence the disordering of the vortex ordered solid is apparently always a first-order transition. We note that $B_{SS}(T)$ being a phase transition line allows one to assert that the free energies of the two solid phases are equal along this line, and they satisfy inequalities of opposite signs as the $B_{SS}(T)$ line is crossed. Our conclusion that this line corresponds to a first-order phase transition implies, in addition, that the entropies of the two solid phases viz. the 'ordered' Bragg glass and the 'disordered' vortex glass, are unequal along this line. The shape of this combined transition line, however, gives rise to many interesting possibilities, including two Kauzmann points [8]. In the present (*B*–*T*) phase diagram of YBCO (Fig. 2) we define these Kauzmann points as the points where the slope of B_{SS} changes its sign—once around 50 K and then at around 75 K.

At a Kauzmann point the entropies of the disordered and ordered state are equal. While in the context of (molecular) liquid–glass transition the liquid is frozen into a glassy state at a temperature $T_G > T_{\mathrm{Kauzmann}}$ to avoid entropy crisis below T_{Kauzmann}, a Kauzmann point can actually be reached in the pressure (*P*)–temperature (*T*) phase diagram of ^{3}He. It leads to the apparently anomalous situation where the solid entropy is higher than the liquid entropy [3]. This is of course now understood in terms of the (nuclear) spin contribution to entropy which due to Pauli exclusion principle is less for the liquid than for the solid. At very low temperatures the spin contribution dominates over the structural contributions, and thus the entropy per atom of the solid is greater than the liquid. Experiments also indicate that poly(4-methylpentene-1) exhibits a pressure maximum, hence a Kauzmann point along its melting curve (see Ref. [8] and references therein). The interpretation of this observation becomes complicated because of the appearence of an additional phase (Ref. [8]).

With this information we now attempt to rationalize the entropic relations between various phases in the vortex matter phase diagram of YBCO. Between two Kauzmann points (50 K,($\approx$)1.1 T and 72 K,($\approx$)2.1 T) in the *B*–*T* phase diagram of YBCO, the $\mathrm{d}B_{SS}/\mathrm{d}T$ has a positive slope; and from the Clausius–Clapeyron relation this will indicate a negative entropy change across this first-order transition line. (The implicit assumption here is that the vortex glass or liquid phase is denser than the ordered vortex solid and the equilibrium magnetization shows a positive jump at the phase transition point as in BSCCO.) Negative entropy leads to the paradoxical situation where the ordered vortex solid has larger entropy than the disordered vortex solid. The name 'ordered vortex solid' itself indicates that it is structurally more ordered. Hence, as in the case of solid ^{3}He the extra entropy needs to be attributed to some additional degrees of freedom.

It has been argued that the non-monotonic behaviour of the transition line as well as the entropy paradox in BSCCO are actually manifestations of change in driving mechanism of the phase transition, namely a disorder driven phase transition at low temperatures to thermally driven phase transition at high temperatures (Ref. [1]). At low temperatures with increase in field the elastic energy decreases relative to pinning, leading to a transition to a disordered phase when the two energies become comparable. Thermal smearing of pinning potential progressively reduces the pinning energy at intermediate temperatures leading to an upturn in the transition line, although the transition remains disorder-driven [28]. In the same vein as in BSCCO (Ref. [1]) it can be argued that in the disordered vortex solid the flux-lines or vortices wander out of their unit cells and become entangled. However, their fluctuations are small on short time scales. In contrast, in the ordered vortex solid there is no large scale wandering of the vortex and no entanglement. But the effect of thermal fluctuations within the unit cell is comparatively large, which in turn results in larger entropy.

There can, however, be other possible sources of extra entropy. An important aspect which is not commonly addressed in the studies of vortex matter is the electronic structure of the vortex cores. The existence of bound quasi-particle states in the normal vortex cores of a conventional superconductor has been predicted in early sixties by Caroli et al. [29] but was not established experimentally until late eighties when Hess et al. [30] observed tunneling spectra in the vortex core of $NbSe_2$ consistent with localized quasi-particle states. With the arrival of HTSC the questions are now asked regarding the low energy physics associated with a vortex core, nodal structure (associated with the proposed $d_{x^2-y^2}$-wave pairings) and quasi-particle transfer between vortices, which will ultimately govern the physical properties of the vortex states. While localized quasi-particle states inside the vortex core have now been observed in YBCO (Ref. [31]), an interesting gap like structure at the Fermi level is found at the centre of the cores of BSCCO which scales with the superconducting gap [32]. Then there exists a class of theories which predicts that magnetism may be induced near or inside the cores of vortices by the application of a magnetic field [33–37]. Neutron experiments on $La_{2-x}Sr_xCuO_4$ suggest that while at optimal doping individual vortices are associated with enhanced low frequency antiferromagnetic fluctuations, the vortex state acquire static long-ranged antiferromagnetism in the underdoped sample [38]. Very recently evidence of static magnetism in the vortex cores of underdoped $YBa_2Cu_3O_{6.5}$ has been provided through muon spin rotation experiment [39].

With these information on the quasi-particles in the vortex cores of the HTSC materials, one can probably bring the analogy between the P–T diagram of ^{3}He and vortex matter phase diagram a bit closer. In an ordered vortex solid the quasi-particles in the individual vortex core will act independently as the spins associated with individual atoms do in ^{3}He solid. In the entangled disordered vortex state the quasi-particles are likely to be more correlated, and hence their contribution to the entropy is reduced.

5. Conclusion

We conclude that the vortex 'ordered solid' to 'disordered solid' transition line in YBCO is probably a first-order transition line. Combining with the earlier results on high temperature low field melting line we suggest that the destruction of the ordered vortex state in YBCO takes place along a unified first-order transition line. The non-monotonic nature of this transition line suggests the existence of two Kauzmann points at around 75 and 50 K. To explain the entropy crisis between these two Kauzmann points, sources for additional degrees of freedom are needed to be identified. Competition between elastic energy and pinning energy in the temperature region of interest and its consequence on the thermal contribution of entropy may be one such source. The other possibility related to the correlation between quasi-particles in the individual vortex cores is also discussed. This latter possibility gains weight from the recent evidence of static magnetism in the vortex cores of underdoped YBCO [39]. From the experimental studies on both YBCO and BSSCO there is no indication as yet of the vortex solid–solid transition line ending in to a critical point, and in order to avoid further entropy crisis the slope of this first order transition line below 50 K needs to be finite and reaching zero value at $T = 0$ K only. In ^{3}He phase diagram the $T = 0$ point is a Kauzmann point but without any entropy crisis since both the ordered and the disordered vortex phase have zero entropy at $T = 0$ K. This need not necessarily be the case in the vortex matter of YBCO and BSSCO where in contrast with ^{3}He, the disordered phase has higher entropy at low temperatures, and this disordered phase will be relatively more susceptible to zero-point vibration at $T = 0$ K.

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