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Order-disorder phase transition in $NbSe_2$: Absence of amorphous vortex matter

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Recent studies have proposed that the peak effect in the critical current (a dynamic property) is a consequence of an order-disorder phase transition in the vortex system. We have made magnetic decorations of vortex structures in samples with both phases. It is found that the structural symmetry of the disordered phase is polycrystalline rather than amorphous and that there is no obvious correlation between the topology of the vortex structure and the enhancement of the critical current in the disordered phase.

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The peak effect in the critical current J_c of low-temperature superconductors as well as the second peak observed in the magnetization loops of high-temperature superconductors have triggered a resurgence of experimental and theoretical research that suggests a unified reinterpretation of the H-T phase diagram.

As indicated in a recent review by Giamarchi and Bhattacharya[1] a unified phase diagram for low- and high-temperature superconductors could provide a general picture of the influence of disorder on the thermodynamic properties of the superconducting state: The peak effect as well as the second magnetization peak are suggested to be the signature of a phase transition between ordered and disordered vortex phases.[1–13] While there is consensus on the nature of the ground state of the ordered phase as that described by a quasi-long-range-ordered Bragg glass,[2,14] the topology of the disordered state remains unclear.[1,5,10,13]

The H-T phase diagram of $NbSe_2$ is the paramount example of a superconductor where the peak effect determines a line $H_p(T)$ separating the ordered vortex phase (OP) (low critical current) from the disordered vortex phase (DP) (high critical current). Although it has been often claimed a reentrance of the DP at low fields, $H_p(T)$ varies from sample to sample and the reentrant disordered phase is not always experimentally detected.[6,7,11,13] The increase of J_c associated with the peak effect is usually attributed to the presence[1,3] of a solid amorphous vortex structure (DP) or a pinned vortex liquid[3] at fields close to $H_{c2}(T)$.

Electrical transport measurements using Corbino contact configuration in Fe-doped $NbSe_2$ provided[9] insight into the problem. The sharp change in J_c at the peak gave support to the existence of a possible first-order phase transition. The usually observed broad increase of the critical current as determined by a four contact strip configuration was attributed[8,9,11] to the penetration of a disordered vortex structure. Corbino transport measurements in Fe-doped $NbSe_2$ showed[9] a clear reentrance of the DP at fields of the order of 1000 Oe.

A recent study[13] of the correspondence between J_c and the vortex topology in real space, as observed by magnetic decorations in pure and Fe-doped $NbSe_2$, has shed doubts on the proposed amorphous nature of the disordered phase. In order to establish the topology of the DP, it would be desirable to decorate a sample that shows the reentrant disordered phase at fields and temperatures where the decoration experiment can be made.

In this work we make a comparative analysis of the vortex structure by Bitter decoration of three $NbSe_2$ samples: A pure $NbSe_2$ sample with no detectable reentrance of the $H_p(T)$ line and two Fe-doped samples, one with no detectable reentrance and the other is that of Ref. 9 where a reentrance close to 1000 Oe was reported.

The experiments were performed on pure $NbSe_2$, sample A, with $T_c = 7.02$ K and two Fe-doped single crystals, sample B with $T_c = 5.8$ K, and sample C (the one used in Ref. 9) with $T_c = 5.7$ K. The H-T phase diagrams of Fig. 1 obtained by Corbino measurements are those reported in Ref. 13 for samples A and B and in Ref. 9 for sample C.

It is known that the behavior of the critical current in the vicinity of the peak effect is sensitive to unspecified material properties. Thus, the $H_p(T)$ line varies not only with Fe doping but even between samples with the same nominal composition. This is clearly seen in Fig. 1: Sample A has a peak effect down to 25 Oe and no reentrance[13] at lower fields, sample B presents a peak effect down to 250 Oe with no reentrance detected for lower fields,[13] and sample C shows a clear reentrance of $H_p(T)$ at 1.2 kOe down to 4.2 K, as reported in Ref. 9.

Magnetic decoration was used to visualize the vortex structure obtained following three different procedures: cooling the sample in the presence of a field (FC), applying the field after cooling in zero field (ZFC), and rotating the field after FC (FCR), experiments.[15] Magnetization measurements indicate that the field B becomes frozen in close to $H_{c2}(T)$ with FC $NbSe_2$ crystals. Moreover, magnetic decoration shows that the frozen B is uniform throughout the sample (the decoration resolution establishes an upper limit for the

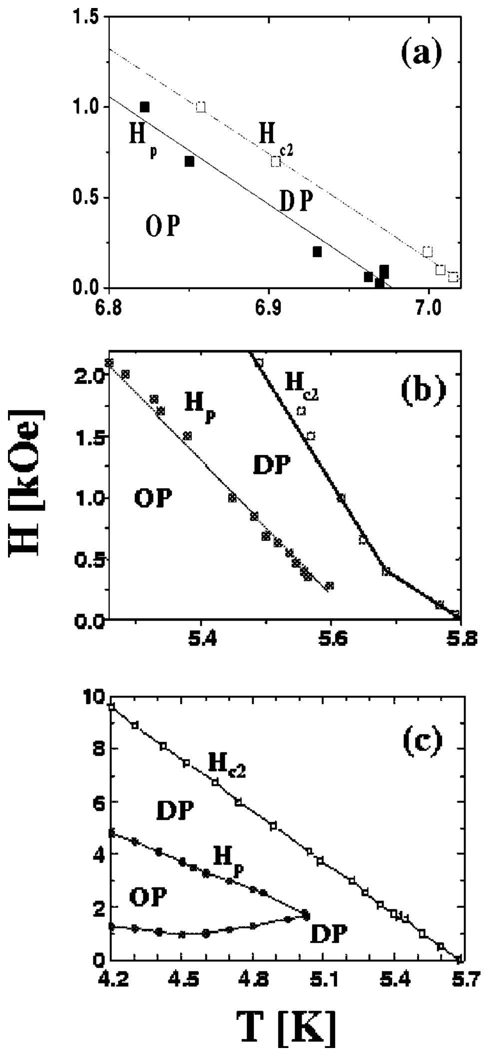

FIG. 1. H-T phase diagrams from Corbino transport measurements. $H_p(T)$ is the boundary between the OP and DP. $H_{c2}(T)$ is the upper critical field. (a) Sample A (pure $NbSe_2$) has a peak down to 25 Oe. (b) Sample B (Fe-doped $NbSe_2$) has a peak down to 250 Oe. (c) Sample C (Fe-doped $NbSe_2$) has a reentrance of the DP below 1.2 kOe.

circulating current density of 10 A/cm^2). As a result, the structure observed by FC is that of the lattice frozen in close to H_{c2} in the DP above $H_p(T)$.

The Delaunay triangulation of magnetic decorations performed in FC experiments at 36 Oe and 3 K is shown in Fig. 2. In Fig. 2(a) the vortex structure of sample A is depicted. Figure 2(b) shows the structure in sample B when FC in a field at which transport measurements[13] do not detect a peak effect. Figure 2(c) shows the vortex structure of sample C when FC within the reentrant DP. The results of samples A and B are similar to those reported in Ref. 13.

Figure 2 shows that contrary to the assumed amorphous structure associated with the disordered region as determined from critical currents the observed vortex topology in this phase is the polycrystalline structure typically found[16–18]at low field. Moreover, in spite of the different critical temperatures and the qualitative differences in the $H_p(T)$ lines, all the structures are similar with nearly the same percentage of vortices in topological defects. Most of the defects are associated with the change of crystalline orientations between grains (GB) and the rest are associated with isolated edge dislocations (I), see Table I. The equivalent FC vortex topology in all samples supports the idea that the observed structures reflect the lattice configuration in the DP, frozen in close to H_{c2}, since otherwise a markedly different structure would be expected in sample C where the DP exhibits the reentrant behavior. The FC data makes clear that the topology of the DP does not correspond to an amorphous structure.

As discussed above, FC decorations capture the vortex structure close to H_{c2} and therefore the topology of the OP would not be accessible by these experiments. On the other hand, the proposed OP - DP phase transition has been associated with a qualitative change in the lattice structure. Such structural changes involve macroscopic displacements of vortices which require forces larger than critical.

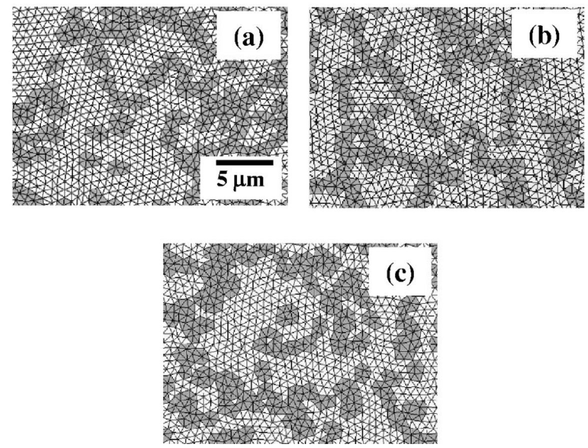

FIG. 2. Delaunay triangulations of the vortex structure obtained after FC at 36 Oe. Gray regions show topological defects. (a) Sample A structure after FC through the $H_p(T)$ line. (b) Sample B structure after FC in a field where no peak in J_c is detected. (c) FC vortex structure of the reentrant DP in sample C. Decorations were performed at 3 K.

TABLE I. Percentage of vortices in topological defects in grain boundaries (GB) and in isolated dislocations (I) for samples A–C when FC at 36 Oe, ZFC at the same field and at 4.1 K, and FCR at different temperatures.

Sample	Defects	FC	ZFC	FCR		
				3.5 K	**4.1K**	
Sample A	GB	14.4%	12.7%	3.1%	0%	
	I	2.7%	2.9%	2.1%	2.3%	
Sample B	GB	16.0%				
	I	1.6%				
				4.1 K	**4.6 K**	**5.0 K**
Sample C	GB	15.7%	11.0%	5.8%	3.9%	3.1%
	I	2.1%	2.7%	2.3%	2.0%	2.3%

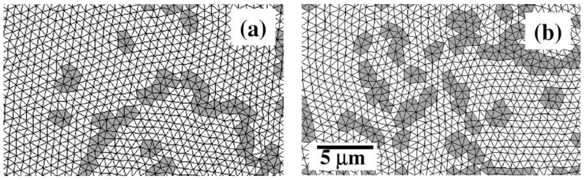

FIG. 3. Delaunay triangulations of vortex structures obtained by ZFC experiments. The magnetic field of 36 Oe was applied at 4.1 K and decorations were made at 3 K. (a) Sample A structure in the OP. (b) Sample C structure in the reentrant DP.

In order to verify whether there is a qualitative topological change induced by the displacement of vortices, we decorated samples A and C after ZFC at 4.1 K and 36 Oe. The obtained vortex structures show gradients[18] of B close to the sample edge, as a result of currents induced when applying the field. Despite this, the bulk of the samples depicts a roughly homogeneous field that allows the observation of the vortex structure of the ZFC state, see Fig. 3. Both the gradient of B and the presence of vortices in the whole sample demonstrate that vortices have been displaced by forces induced by currents larger than the critical. Figure 3 makes evident that there is no qualitative vortex topological difference between the structure obtained by ZFC in the OP in sample A and that in the DP in sample C; both are polycrystalline with almost the same grain size and percentage of vortices in grain boundaries, see Table I.

It can be argued that the overall polycrystalline nature of sample A is due to a mixture of DP and OP phases when ZFC. The mixture could be induced either by a contaminated phase introduced through sample edges[8] or as a result of the disorder induced by plastic motion.[19]

In order to overcome the limitations of ZFC we have used an efficient method of annealing the vortex lattice by FCR experiments,[15] where the vortex structure is ordered by a rapid field rotation. We have applied the FCR vortex crystal growing method to samples A–C in regions of the phase diagram corresponding either to the OP or DP. We have performed FCR experiments with a c-axis field of 36 Oe and an in-plane component $H_T = 72$ Oe, added and removed suddenly at 4.1 K. The decorations were made at 3.5 K for all samples.

The FCR was applied within the OP in sample A, the DP in sample C, and in a region where no reentrance of DP has been detected by critical current measurements in sample B, see Fig. 1. The results are shown in Fig. 4; the increase of the crystalline order as compared with the FC case is evident. In sample A the grain boundaries are suppressed over the entire sample. Although the grains in samples B and C are smaller than the size of the respective samples, they involve on average a number of vortices nine times larger than in the FC case.

From Fig. 4 it could be deduced that the FCR vortex structures correspond to the genuine vortex topology of the equilibrium state: a single crystal in the OP region of sample A and a polycrystalline structure in the DP region of sample C. On the other hand, a critical analysis of the results indicates that it is difficult to know whether the difference between the vortex topology after FCR in the OP and DP should be at-

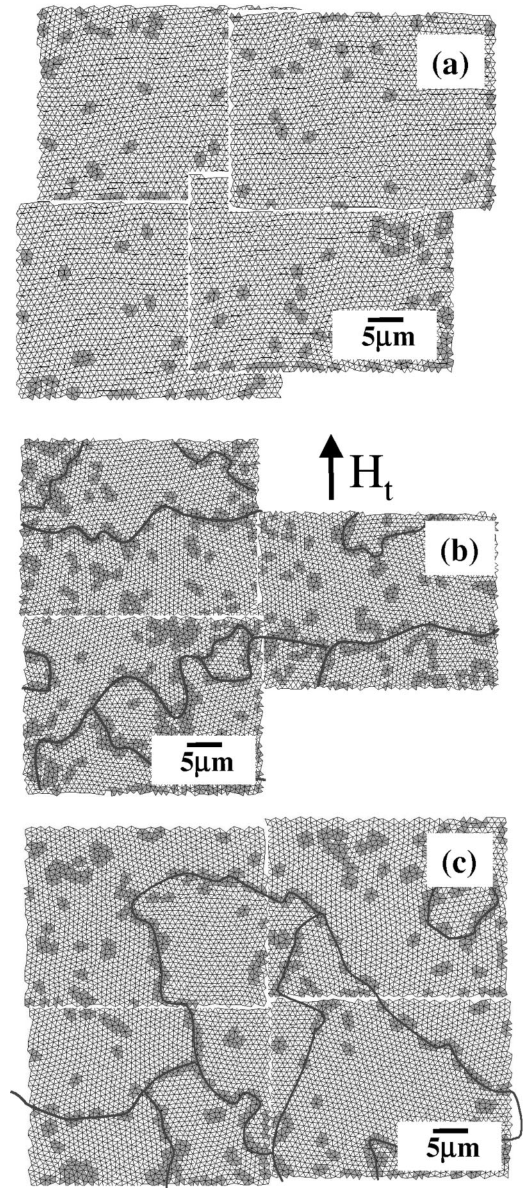

FIG. 4. Delaunay triangulations of vortex structures after FCR process with a field parallel to the c axis of 36 Oe and an in-plane component $H_T = 72$ Oe. The rotation was carried out at 4.1 K and decorations were made at 3.5 K. Results for (a) sample A, (b) sample B, and (c) sample C.

tributed to two different equilibrium states or to a limited efficiency of the annealing technique when applied to samples with different critical currents. This is supported by FCR experiments at 3.5 K in sample A where it was not possible to fully remove the grain boundaries and where the critical current is known to be larger[20] by about 20% as compared to that at 4.1 K. The results suggest that the ability to create single crystals applying FCR in sample A is limited by nucleation and growth processes in the presence of an effective viscosity associated with the pinning strength, as suggested in earlier work.[21]

As a result of the previous observation we foresee an experimental method to distinguish whether the polycrystalline structure obtained in sample C is associated with an

TABLE II. Average number of vortices in grains for sample C when FC at 36 Oe and FCR at 4.1 K, 4.6 K, and 5.0 K.

FC	FCR	
200	4.1 K:	1700
	4.6 K:	2500
	5.0 K:	4900

equilibrium DP structure or the result of imperfect crystal growth in the presence of pinning. In the first case the FCR at higher temperatures should induce an increase of the percentage of vortices in grain boundaries (decrease of grain size), since the equilibrium density of dislocations is generally expected[10] to increase with temperature. On the contrary, if the grain boundaries are associated with a limitation of the efficiency of the crystal growth technique, the size of the grains should increase when FCR at higher temperatures due to the decrease of J_c with temperature. We have performed FCR at higher temperatures and the corresponding structures were magnetically decorated at 3.5 K. The results of FCR experiments at 4.1, 4.6, and 5 K in sample C show a continuous decrease of the percentage of vortices associated with grain boundaries, see Table I, or equivalently, an increase of the grain size in the magnetic decoration image, see Table II. It should be remarked that the percentage of vortices (3%) in grain boundaries in the FCR experiment at 5 K is much smaller (larger grain size) than that in FC experiments. This indicates that the grain boundaries obtained in FC experiments are associated with the nucleation and growth processes of the crystalline regions.

By means of systematic real-space vortex structure observations we conclude that the vortex topology of the DP, in contrast with the expectations, is not amorphous and, more importantly, most of the detected defects are concentrated in grain boundaries that sourround large crystalline grains containing thousands of vortices. The FCR data obtained in the reentrant DP suggest that the observed density of vortices involved in defects at each temperature sets only an upper limit, and consequently the equilibrium state of the DP could be characterized by an even lower density of defects. As a result it is counterintuitive to associate the low density of dislocations that charaterizes the DP region as the driving mechanism of the sharp increase of the critical current at the OP - DP transition. Therefore, no correlation between the topology of the vortex structure and the enhancement of the critical current in the disordered phase has been found.

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