Ordered, Disordered, and Coexistent Stable Vortex Lattices in $NbSe_2$ Single Crystals

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The peak effect (PE) in the critical current density of type II superconductors has been related to an order-disorder transition in the vortex lattice (VL), but its underlying physics remains a controversial issue. Intrinsic to the PE are strong metastabilities that frequently mask the *stationary* VL configurations. We follow shaking and thermal protocols in $NbSe_2$ single crystals to access these configurations and examine them by linear ac susceptibility measurements that avoid VL reorganization. We identify three different regions. For $T < T_1(H)$, stable VL configurations are maximally ordered. For $T > T_2(H)$, configurations are fully disordered and no metastability is observed. In the $T_1 < T < T_2$ region, we find temperature-dependent stable configurations with an intermediate degree of disorder, possibly associated with the coexistence of ordered and disordered lattices throughout the PE.

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The behavior of systems under the influence of both thermal and quenched disorder has been an issue of intense research over the past 50 years in different areas of condensed matter physics. Quenched disorder can broaden first order phase transitions inducing phenomena such as phase separation (under active study in the context of manganites, for instance) and if sufficiently strong can even change completely the properties of this system, producing the appearance of new phases (as is the case in many glassy states) [1].

In this context, the vortex lattice (VL) in superconductors provides model systems where elastic and pinning interactions together with thermal fluctuations compete. A fingerprint of the complex behavior in this system is the peak effect (PE), an anomalous nonmonotonic dependence of the critical current density J_C with both temperature and magnetic field [2]. The origin and nature of this behavior are still controversial issues.

Phenomenological pictures based on an order-disorder (O-D) transition from a quasiordered Bragg glass (BG) [3] to a disordered phase with proliferation of topological defects [4] explain a broad number of related experimental results. However, the underlying physics of the transition, the structural symmetry of the disordered phase [5], and its connection with a clean limit melting transition [6,7] are issues that remain unsolved. While an increasing amount of experimental evidence [8] suggests that the nature of the PE could depend on the various materials, the marked metastability and history effects are common facts reported both in low [9–11] and in high T_c [12] materials.

In traditional superconductors, neutron diffraction experiments show a clear change in the structure factor lattice at the PE, indicating an O-D transition [13]. In $NbSe_2$, metastable coexisting regions with different J_c have been directly observed [14]; this fact as well as transport measurements in the Corbino geometry [11] and experiments of magnetization assisted by a shaking ac field [10] suggest a first order transition where the ac magnetic field assists an equilibration process from a supercooled disordered metastable phase to an ordered stable BG phase.

PE and history effects are often studied by means of transport experiments. In standard transport techniques, vortices undergo some type of current-induced reorganization, and therefore the original vortex lattice configuration (VLC) is not accessible. To overcome this problem, Xiao *et al.* [7] have developed a sophisticated ultrafast transport technique and have shown the existence of an enlarged crossing phase boundary between ordered and disordered phases in $NbSe_2$ crystals. A well-known and appropriate technique to measure the pinned VL response without external disturbances is ac susceptibility restricted to the linear Campbell regime [15], where a very small ac field h_a superimposed to the dc field H is applied, forcing vortices to perform small (harmonic) oscillations inside their effective pinning potential wells.

In this Letter, we explore quasistatic VLCs in the Campbell regime in $NbSe_2$ single crystals in the vicinity of the PE following different thermal, magnetic, and dynamical histories. Essential to our work, we avoid any measurement-induced VL reorganization. We are able to access the corresponding *stable* configuration at each temperature (field) by applying a shaking ac field. For the first time, the pinning potential curvature of stable quasistatic VLCs is measured throughout the PE, resulting in a clear identification of low temperature maximally ordered, high temperature maximally disordered, and stable configurations with an intermediate degree of disorder at intermediate temperatures. In the last case, the stable VLCs are separated by energy barriers, possibly related to surfaces between coexisting ordered and disordered VL domains. We can also observe the evolution of the pinning potential with temperature in *metastable* VLCs and confirm the spontaneous ordering in cooling processes [16].

Results shown here correspond to a $NbSe_2$ single crystal [17] of approximate dimensions $(0.5 \times 0.5 \times 0.03)$ mm^3, with $T_c = 7.30$ K (defined as the midpoint of the ac susceptibility linear transition at $H = 0$) and $\Delta T_c = \pm 0.02$ K. Ac susceptibility has been measured with a homemade susceptometer based on the mutual inductance technique with a system coil similar to that described in Ref. [18]. All fields are parallel to the c axis of the sample.

In the Campbell limit, the inductive component of ac susceptibility χ' is determined by the geometry and the dimensionless parameter λ_R/D (λ_R is the real penetration depth, and D is a characteristic sample dimension). In this regime the imaginary penetration depth $\lambda_I \ll \lambda_R$. The curvature of the effective pinning potential well is the Labusch constant α_L that can be numerically estimated from $\lambda_R = (\lambda_L^2 + \phi_0 B / 4\pi\alpha_L)^{1/2}$, where λ_L is the London penetration depth. To estimate λ_R, we use a numerical calculation for a disk in a transverse ac field. The dimension coils allow us to approximate the sample to a magnetic dipole (for references and details of the numerical procedure, see Ref. [18]).

Figure 1 shows typical $\chi''(T)$ (a) and $\chi'(T)$ (b) field cool cooling (FCC) and field cool warming (FCW) curves in the linear ($h_a = 0.025$ Oe, black curve) and nonlinear regimes ($h_a = 0.32$ Oe, gray curve) at $H = 320$ Oe. The temperatures at which χ' shows a maximum or a minimum ($T_p^{\rm on}$ and T_p in Ref. [19]) slightly depend on h_a or history. Above a temperature $T_2 \gtrsim T_p$, FCW and FCC curves merge. Below T_2, $\lambda_I \lesssim 0.1\ \lambda_R$ (dissipation is very small) and χ' is frequency-independent in the kilohertz range. Both facts are evidence for a linear Campbell regime. In the nonlinear regime, where vortices are forced out of their pinning sites, both FCC and FCW curves display PE, whereas in the linear Campbell regime, it appears only in the warming process. As the PE is a signature of an O-D transition, its absence in the FCC Campbell regime indicates that the VL nucleates at high T and remains trapped in a metastable disordered and strongly pinned configuration. On the other hand, even if ac-induced currents are not able to assist the VL, the system warms with a lower pinning potential (larger penetration depth). In this framework, the hysteresis displayed in the Campbell regime must imply some kind of spontaneous reordering at low temperatures. This ordering during FCC was recently observed by using a time-resolved transport technique [16]. In the inset in Fig. 2, $\chi'(T)$ recorded in various warming processes from different low temperatures is shown. A lower initial temperature results in a less pinned (more ordered) VLC. We have tested that the warming curves are independent of the cooling or warming rate and are identical if the cooling process is performed without measurement.

This spontaneous ordering at low temperatures can be enhanced by applying a large shaking ac field. This shaking field allows the system to explore the free-energy landscape and to reach a more ordered and stable VLC. This feature is illustrated in the main panel of Fig. 2. The result of the FCW process from 4.2 K shown in the inset (gray line curve in both panels) is compared with various warming curves (symbols) recorded after shaking the lattice at

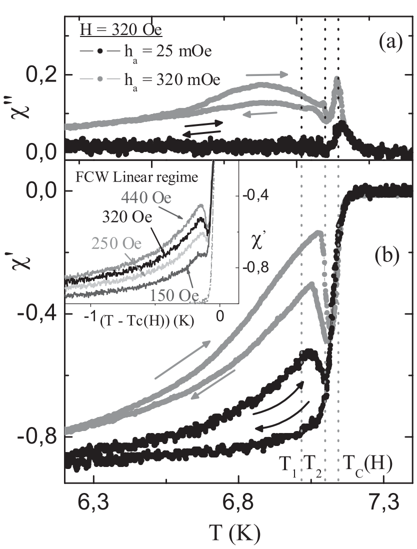

FIG. 1. Typical $\chi''(T)$ (a) and $\chi'(T)$ (b) FCC and FCW curves in the linear (black curves) and nonlinear regimes (gray curves) measured at $f = 30$ kHz. Arrows indicate the direction of temperature variation. T_1 and T_2 limit the T regions of stable ordered and disordered phases (see text). Inset: Linear FCW curves at various dc fields plotted as a function of $[T - Tc(H)]$. An abrupt PE develops for $H \geq 200$ Oe. The zero dc field transition is also shown.

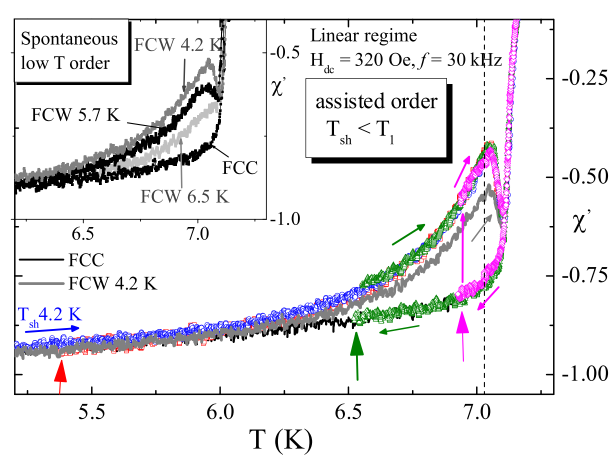

FIG. 2 (color online). Inset: $\chi'(T)$ recorded in various warming processes from different T: There is a progressive spontaneous ordering in the cooling process. Main panel: FCC and FCW (4.2 K) processes (gray lines) compared with curves (various symbols) recorded before and after shaking the VL at different temperatures $T_{\rm sh}$ (large vertical arrows). Vertical (magenta) line remarks the huge χ' change after shaking. All shaken VLCs collapse to the same reversible curve. Small arrows indicate the change in temperature with time.

different temperatures T_{sh} (large vertical arrows). The following process has been performed: The system has been measured in the linear regime during the FCC procedure and stabilized at T_{sh}. Then the measurement was interrupted, and a sinusoidal shaking ac field of 3.2 Oe–30 kHz was applied during 30 s. The measurement (h_a = 25 m Oe) was resumed in a warming procedure starting from T_{sh} leading to a drastic decrease of pinning (vertical color lines in the figure). The various symbols are hard to distinguish because all warming procedures collapse to the same curve. This curve has remarkable characteristics below a temperature $T_1 < T_p^{on}$ (dotted line in the figure): First, it is reversible; second, after any additional shaking, with any waveform or amplitude, the system remains in this configuration [20]. These features constitute a first important result: Below T_1, the stable VLCs (attained via smooth temperature changes) are continuously connected in a minimum of the free-energy landscape. These are the less pinned (more ordered) VLCs at each T. This picture is consistent with a stable ordered BG below T_1, where changes in T produce only elastic (reversible) deformations.

The description changes completely when $T_1 < T < T_2$: The more ordered VLCs become unstable and the PE develops. Again, the system does not reach spontaneously a stable VLC: Warming (cooling) leads to metastable over ordered (disordered) configurations. In every case, a large shaking field assists the system in accessing a stable VLC. The qualitative difference observed above T_1 is that, once a stable VLC has been reached, any smooth temperature variation results in new metastable states: The stable configuration at the new temperature is reached only through shaking; consequently, no reversibility is possible. This remarkable feature is one of the central issues of this Letter and is illustrated in Fig. 3, where χ' values in the linear regime corresponding to the stable VLCs at various T are plotted in large symbols. The complete experiment shown in Fig. 3 is the following: First, a warming process (small black dots) was performed, stabilizing temperature successively at very small intervals ΔT_{sh} [21]. At each T_{sh} (indicated by black large vertical arrows), the measurement was interrupted, and the same shaking protocol described above was performed, in a way to reach the stable VLC (open triangles). At fixed temperature, additional perturbations do not modify the response, and the system remains in this stable configuration. Then the measurement on warming was resumed (small black dots) until the next T_{sh} was reached. As T increases, the corresponding stable VLC is more disordered.

Once $T > T_2$ was reached, a cooling process (small gray dots) was performed, and a similar protocol was followed, shaking the system at the same temperatures (gray vertical arrows). Notice that $\chi'(T_{sh})$ obtained after shaking in warming (hollow triangles) and cooling (gray circles) is the same (within experimental resolution). This central

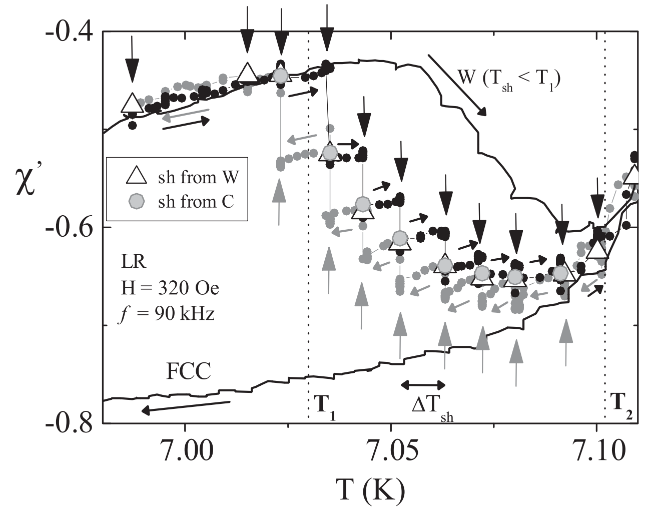

FIG. 3. Warming (W) and cooling (C) experiment between T_1 and T_2. Vertical down (up) arrows identify the T_{sh} in the W (C) process. At each T_{sh}, the lattice is shaken to find the stable VLC (large open triangles in W and large gray circles in C). The stable VLC warms (cools) in a metastable VLC (small dots). The more ordered and disordered curves are also shown as a black line for reference.

result supports the stability of VLCs with intermediate order and rules out the possibility that they arise from a harder metastability or slow dynamics. The more ordered (warming curve after shaking at $T_{sh} < T_1$) and the more disordered FCC curves are shown for reference as a black line.

To quantify the pinning strength on the various VLCs, the effective Labusch parameter $\alpha_L(T)$ corresponding to each VLC has been estimated from the χ' values in the linear regime at 90 kHz in the region where $\lambda_I \ll \lambda_R$. The result is shown in Fig. 4(a). Black curves correspond to metastable VLCs evolving in FCC and FCW processes, whereas the gray curve corresponds to ordered VLCs attained by shaking at 6.4 K. Hollow circles show α_L corresponding to stable VLCs between T_1 and T_2 extracted from Fig. 3. It can be seen that all of the $\alpha_L(T)$ curves approach at low T. At $T > T_1$, the ordered VLCs (gray curve) become metastable. The α_L corresponding to the stable VLC increases more than 5 times in the PE, indicating a huge increase of disorder, qualitatively different to the smooth and monotonically increasing $\alpha_L(T)$ observed in $YBa_2Cu_3O_7$ crystals [8,20]. The ordered (reversible) gray curve below T_1 corresponds to a BG, free of dislocations, and the black (reversible) curve above T_2 corresponds to a completely disordered phase. The stable character of the VLCs with intermediate degree of disorder for $T_1 < T < T_2$, together with additional evidence of coexisting domains in $NbSe_2$ [14], allows us to propose a scenario of equilibrium phase separation. In this framework, the metastability would arise from an exceeding proportion of ordered or disordered domains. To give a

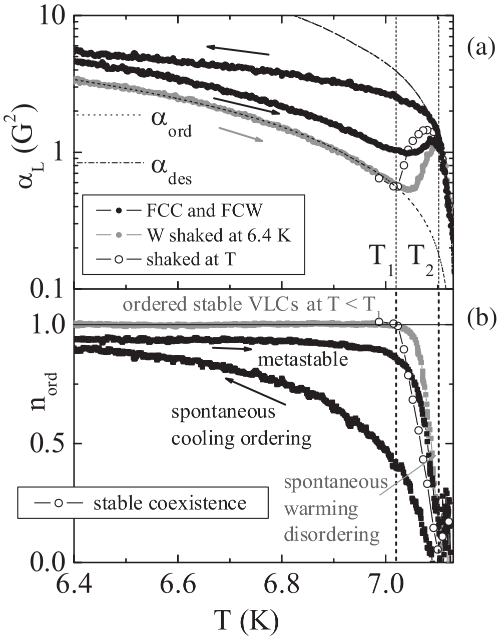

FIG. 4. (a) $\alpha_L(T)$ in log scale corresponding to the various stable and metastable VLCs. $\alpha_{\rm ord}$ ($\alpha_{\rm dis}$) is a fitting curve that extrapolates $\alpha(T)$ in a completely ordered (disordered) VL. (b) Estimated proportion of ordered phases in each VLC. Arrows indicate the change in temperature with time.

rough estimate of the proportion of ordered phase, we proceeded in the following way: We extrapolated the ordered Labusch parameter $\alpha_{\rm ord}(T)$ to zero at $Tc(H)$ [dotted line in Fig. 4(a)], and we proposed $\alpha_{\rm dis}(T) = 7\alpha_{\rm ord}(T)$ to fit the black curve above T_2 [dashed-dotted line in Fig. 4(a)]. We then used the calculated $\alpha_L(T)$ to estimate the proportion of ordered phase $n_{\rm ord}(T) = [\alpha_{\rm dis}(T) - \alpha_L(T)]/[\alpha_{\rm dis}(T) - \alpha_{\rm ord}(T)]$ corresponding to each VLC. The result is shown in Fig. 4(b).

We arrive then at the following scenario. There are three different temperature regions along the PE. For $T < T_1(H)$, stable VLCs are maximally ordered and accessible by the application of a shaking ac field. We identify these ordered stable VLCs as a BG, continuously connected by elastic deformations in a minimum of the free-energy landscape. The robust metastable states below T_1 show spontaneous ordering at low T as elastic interactions overcome pinning forces, although they remain separated from the BG by a finite barrier in all of the experimental range, down to 4.2 K. For $T > T_2(H)$, configurations are fully disordered and no metastability is observed. We identify $T_2(H)$ with $Ts(H)$, the spinodal line in Refs. [7,19]. The evolution from the ordered BG to the high temperature disordered phase, in the small $T_1 < T < T_2$ region, occurs by a gradual increase in the proportion of disorder. The equilibrium VLCs are accessed only from overordered or overdisordered configurations by a shaking ac field that provides sufficient energy for plastic (irreversible) deformations to access the stable VLC. Plastic changes imply the creation or annihilation of dislocations, probably related to the movement of domain walls in a scenario of equilibrium phase separation. In conclusion, by measuring for the first time the pinning potential curvature of stable quasistatic VLCs, avoiding vortex lattice reorganization, we are able to present a consistent picture that describes the vortex lattice physics in the PE region in $NbSe_2$ single crystals.

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