Observation of the Vortex Lattice Spinodal in $NbSe_2$

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Metastable superheated and supercooled vortex states in $NbSe_2$ crystals were probed with fast transport measurements over a wide range of field and temperature. The limit of metastability of the superheated vortex lattice defines a line in the phase diagram that lies below the superconducting transition and is clearly separated from it. This line is identified as the vortex lattice spinodal, and is in good agreement with recent theoretical predictions by Li and Rosenstein [Phys. Rev. B **65**, 220504 (2002); cond-mat/0305258]. By contrast, no limit of metastability is observed for the supercooled disordered state.

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Systems of many interacting particles often exhibit metastability and irreversible behavior that betray the presence of local minima in the free-energy landscape. In the vicinity of a first-order transition, these local minima can trap the system in a metastable superheated or supercooled, "wrong" phase [1]. The metastable region extends up to the spinodal, a line in the phase diagram that demarcates the limit of stability with respect to fluctuations toward the thermodynamically stable state. Vortices in superconductors provide a striking example of such a system with metastability manifesting itself in supercooling [2,3] and superheating [4] as well as in more surprising ways such as frequency memory [5] and nonlinear dynamics [6–9]. These phenomena appear in conjunction with a sudden increase in the critical current as a function of field or temperature, the so-called peak effect [10], which is attributed to an order-disorder transition in the vortex lattice [11–14]. Although the region of metastability associated with this transition is observed in many experiments, there is until now no evidence of a spinodal line. In this Letter, we report results of fast transport measurements on a superheated vortex lattice that identify for the first time a spinodal line above which the system becomes reversible.

The thermodynamically stable state of a type II superconductor in a magnetic field at low temperatures is a lattice of vortices with quasi-long-range order [15]. As the temperature is raised, the vortex lattice undergoes a first-order transition [16,17] to a stable disordered state. Transport measurements provide a fast and accurate way to determine the degree of order in the vortex lattice, even for very small samples where techniques such as neutron scattering cannot be used. As shown by Larkin and Ovchnnikov [11], the critical current where vortices first start moving, I_c, decreases with increasing degree of order as characterized by the size of coherent domains. If, however, the measurement of I_c is not sufficiently fast, as is often the case in standard transport techniques, it will reflect a vortex lattice that has undergone some type of current-induced organization rather than the initial state [8] because vortices would have enough time to reorder in response to the driving current. Current-induced organization can result in a more disordered lattice due to "edge contamination" [18] by new vortices entering through a surface barrier at the sample edge [19,20], or in a more ordered lattice due to "motional ordering" when vortices are driven at high velocities [21]. To avoid current-induced organization, we developed a technique that probes the vortex response on time scales shorter than reorganization times and can capture the response of the initial (static) vortex state much like a snapshot. The technique employs a four-probe measurement to monitor the vortex response to an applied current ramp and I_c is defined as the current for which the response reaches 5 μV. The ramps used in most measurements discussed here were fast-current ramps (FCR) with a sweep rate of 200 A/s. The sweep rate controls the degree of current-induced organization. In the case of FCR, the measurement time to obtain a voltage signal within our resolution of $\Delta V \sim 1\ \mu$V is $\tau = \Delta V/(R_{ff} dI/dt) \sim 2\ \mu$s, where R_{ff} is the free flux flow resistance [22]. During this time, the vortex moves less than half a lattice spacing for typical field values used here. By contrast, if the same vortex is probed with a slow-current ramp $\sim$1 mA/s, $\tau \sim 0.4$ s is much longer than the time to traverse the entire sample (typically $\sim$50 ms at 0.25 T and 5 μV) so current-induced organization is inevitable.

The experiments were carried out on three 2H-$NbSe_2$ crystals with critical temperatures $T_c = 5.61$, 7.01, and 6.0 K for samples A, B, and C, respectively. In Fig. 1, we show the temperature dependence of I_c in sample A for vortex states obtained by three methods of preparation: (i) Field cooling (FC), open diamonds, whereby the field is applied prior to cooling through the superconducting

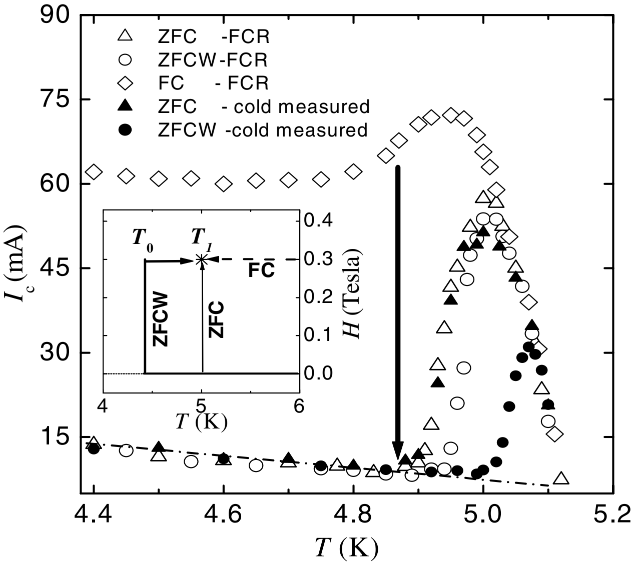

FIG. 1. Temperature dependence of critical currents at $H = 0.3$ T obtained with several preparation and measurement methods. Open symbols—measured at the preparation temperature. Solid symbols—calculated from cold measured critical currents as described in the text. The arrow indicates the decay of the supercooled FC state. Inset: Field-temperature trajectories used in various methods of preparation.

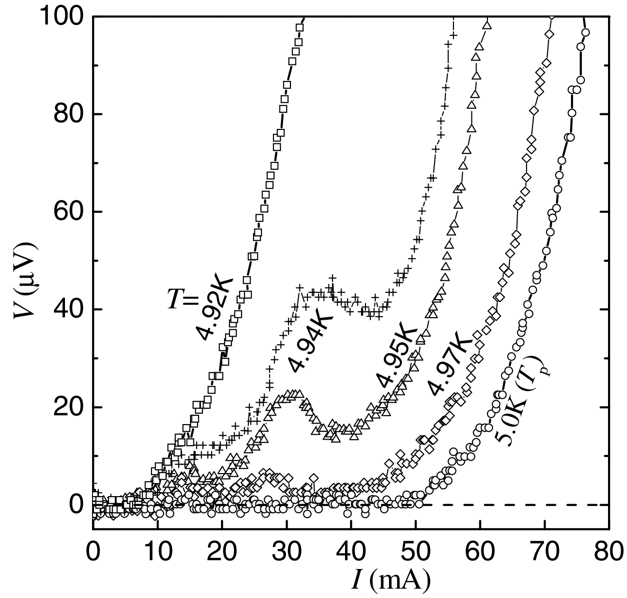

FIG. 2. N-shaped voltage-current curves for the ZFCW lattice at $H = 0.3$ T in sample A close to the peak temperature are a signature of current induced organization.

transition. In this case the final vortex lattice is in a disordered state as indicated by the high values of I_c. At low temperatures the FC state is metastable. Any disturbance causes it to decay into the ordered state as indicated by the arrow. (ii) Zero Field Cooling (ZFC), open triangles, is obtained by applying the field after cooling to the desired temperature. In this case, vortices penetrate from the periphery moving into the sample at high speeds and forming a motionally ordered state with I_c significantly lower than in the FC case. At low temperatures the ZFC state is stable. (iii) ZFC Warm (ZFCW), open circles, entails preparing the ZFC lattice at a low temperature $T_0 = 4.2$ K, and then heating it in the absence of applied current to the target temperature T_1 where it is allowed to thermalize for 2–3 min and then measured with FCR. This process results in a superheated lattice which remains ordered up to a temperature higher than either the ZFC or the FC lattice.

When probing the properties of a metastable state, the measurement process can drive the system into the more stable state. Therefore in order to establish the limit of superheating it is necessary to use noninvasive measurements. Indeed, we found that in spite of using FCR the V-I curves of the ZFCW state in the vicinity of peak effect, shown in Fig. 2, were N shaped which is a signature of current-induced organization [9,20]. Since this indicates that our measurement is too slow in this temperature range, we followed a different procedure: The ZFCW lattice was not probed at T_1 but, after waiting at T_1 for a few minutes, it was cooled back to T_0 and then measured with FCR. The critical currents obtained by this procedure, henceforth referred to as "cold-measured-critical currents" and labeled $I_{cc}(T_1)$, are shown in Fig. 3 (circles). They were used to calculate (as described below) the values of $I_c(T)$ for the ZFCW state in the temperature range where they were not accessible by direct measurement. These calculated values are shown in Fig. 1 by solid circles. For comparison, we also show in Fig. 3 the cold-measured critical currents $I_{cc}(T_1)$ for the ZFC state (triangles) obtained in a similar manner by preparing the state at T_1 and measuring it after cooling to T_0. The

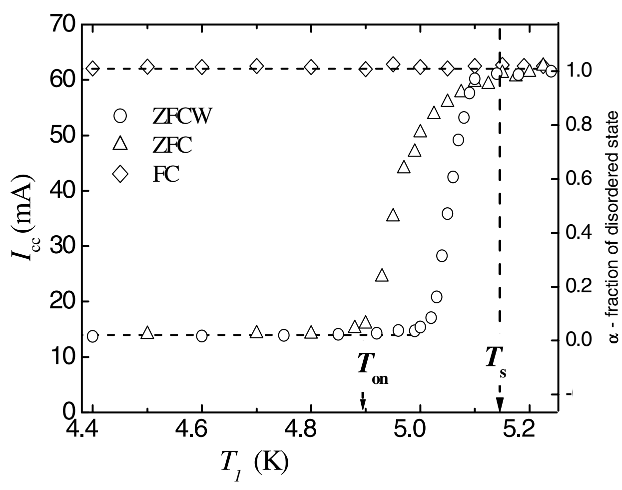

FIG. 3. Cold-measured critical currents at $H = 0.3$ T, for vortex lattices prepared at T_1 and measured at $T_0 = 4.4$ K. T_s is the limit of metastability for the ordered state. For $T < T_{on}$ excursions in temperature do not affect I_{cc}. The right-hand ordinate is the fraction of disordered state along the minimal current cross section as described in the text.

same qualitative behavior is seen in both data sets: at low temperatures, $I_{cc}(T_1) = I_{co}(T_0)$ is constant, where $I_{co}(T_0)$ is the critical current of the ordered state prepared and measured at T_0. Above an onset temperature T_{on}, $I_{cc}(T_1)$ starts increasing and ultimately saturates to the value of the disordered state, $I_{cd}(T_0)$, at a higher temperature $T_1 = T_s$. For $T < T_{on}$, excursions in temperature leave the critical currents unchanged, while excursions beyond it lead to an irreversible increase in $I_{cc}(T_1)$. The saturation temperature, $T_s \sim 5.18$ K, is highest in the ZFCW lattice as expected of a superheated state. We will show below that this temperature corresponds to the spinodal point. In the same figure, we show the data for the FC lattice (diamonds) which remains in a supercooled disordered state at all temperatures.

We attribute these results to the crossing of a phase boundary between ordered and disordered states. In the vicinity of this boundary domains of ordered and disordered phase can coexist as shown previously by Hall probe microscopy [23] and magneto-optical imaging [24]. The critical current in this heterogeneous state is determined by the sample cross section along which the average critical current density is lowest, corresponding to the cross section where the fraction of disordered state is minimal. At the onset of vortex motion, the current density along the minimal cross section takes the value of the local critical current density [25] as determined by the type of domain traversed. It follows that, if α is the fraction of disordered phase measured along this path, the critical current will be $I_c = \alpha I_{cd} + (1 - \alpha) I_{co}$, where I_{cd} and I_{co} are the critical currents in the disordered and ordered phases, respectively. Our data suggests that the disordered domains remain unchanged upon cooling, so that the cooled state is a replica of the state at T_1. Therefore, the cold-measured critical currents correspond to a state where the fraction of disordered state is $\alpha(T_1)$ and $I_{cc}(T_1) = \alpha(T_1) I_{cd}(T_0) + [1 - \alpha(T_1)] I_{co}(T_0)$. Using the measured values of $I_{cc}(T_1)$, $I_{cd}(T_0)$, and $I_{co}(T_0)$, we obtain $\alpha(T_1)$ also shown in Fig. 3. We note that this "two-phase-coexistence" model gives $I_{cc}(T_1) = I_{co}(T_0)$ at $\alpha(T_1) = 0$, even if the sample contains islands of disordered phase, as long as there is a contiguous sheet connecting opposite edges of the sample in which all vortices are in the ordered phase. Such a sheet cannot be found when the disordered phase percolates cutting the sample lengthwise—in this case $\alpha(T_1) > 0$ and $I_{cc}(T_1) > I_{co}(T_0)$. Beyond this point, as $\alpha(T_1)$ increases with temperature, so does $I_{cc}(T_1)$. As long as $\alpha(T_1) < 1$, there are ordered islands embedded in the sample until they disappear at $\alpha(T_1) = 1$, where the entire sample is disordered. For a ZFCW lattice measured with FCR at T_0, the current-induced organization is essentially absent so that the disordered phase can only be nucleated by thermal fluctuations. Therefore, the limit of superheating of the ordered phase coincides with the point where the ZFCW lattice becomes thermally disordered, at T_s. Indeed, for $T > T_s$ we find that the vortex lattice is always disordered, and metastability or hysteresis are absent, regardless of the method of preparation or measurement speed. We conclude that it is not energetically possible for an ordered domain to exist above this temperature, and therefore we identify it with the spinodal point for the superheated vortex lattice in 2H-$NbSe_2$. By contrast, we found no limit of supercooling for the FC state. By repeating the measurements for several field values, we obtained the phase diagram shown in Fig. 4 in terms of the reduced variables $t = T/T_c$ and $h = H/H_{c2}$, where H_{c2} is the upper critical field at $T = 0$ [26]. The spinodal points t_s lie on a line well separated from the critical line t_c and is above the onset t_{on} and peak t_p temperatures for the ZFC lattice.

As a check on the two-state coexistence model, we used the values of $\alpha(T_1)$ to calculate $I_c(T_1) = \alpha(T_1) I_{cd}(T_1) + [1 - \alpha(T_1)] I_{co}(T_1)$, the critical current of the static vortex lattice at temperatures T_1, where it is experimentally accessible with the FCR. The calculated $I_c(T_1)$ for the ZFC state, shown in Fig. 1 (solid triangles), are in close agreement with the directly measured data validating the assumptions in the model. In the same figure, we show the calculated values of $I_c(T_1)$ for the ZFCW state (solid circles) up to temperatures where it is experimentally inaccessible with FCR.

Repeating the experiments for samples B and C produced the same behavior. Both samples revealed a spinodal point for the superheated vortex lattice but no evidence for a limit of supercooling. As shown in Fig. 4, the spinodal points for samples B and C lie on the same

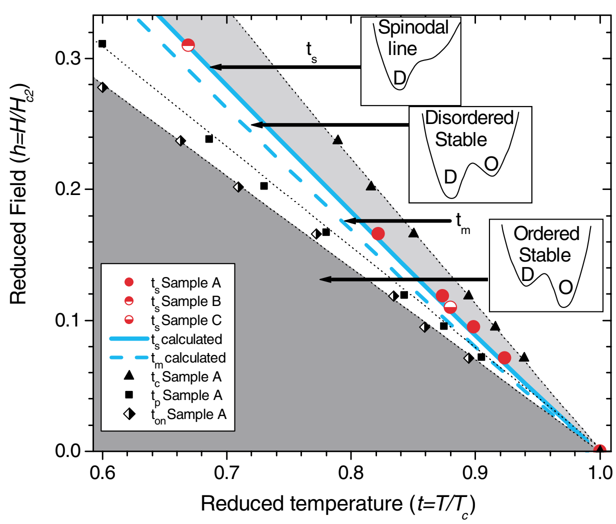

FIG. 4 (color online). The measured limit of superheating for samples A, B, and C (circles), is compared with the calculated LR spinodal (solid line). The calculated melting line (dashed line) lies well above the measured peak (squares) and onset (diamonds) temperatures of the peak effect. The schematic free energy diagrams show the relative depth of the minima of the disordered (D) and ordered (O) phases in each regime.

line as those of sample A even though they are from different batches, with different impurity content.

We compare our data to recent theoretical results of Li and Rosenstein [27] (LR) for the spinodal and melting lines. In terms of the dimensionless scaled temperature, $a_T(t, h) = -[(\mathrm{Gi})^{1/2}\pi/2^{1/2}]^{-2/3}[(1-t-h)t^{-2/3}h^{-2/3}]$, the LR theory gives the melting and spinodal lines at $a_T = -9.5$ and $a_T = -5$, respectively. Here $\mathrm{Gi} = 1/8(k_B T_c/\varepsilon\varepsilon_o\xi)^2$ is the Ginzburg number [27], ξ the zero temperature coherence length, $\varepsilon = (m_{ab}/m_c)^{1/2}$ the anisotropy parameter, $\varepsilon_o = (\Phi_o/4\pi\lambda)^2$ the characteristic vortex line energy, and λ the London penetration depth at zero field and temperature. We used $\varepsilon = 0.3$, $\lambda = 135$ nm [28], and $\xi = 8.8$ nm [26] for sample A ($\xi = 8.2$ nm, 8.3 nm for samples B and C, respectively) to calculate the LR lines for the melting and spinodal shown in Fig. 4. The LR spinodal lines for the three samples merge into the line thickness in the figure. We note that the LR spinodal line practically coincides with our data and is well separated from both the melting line $t_m(h)$ and from $t_c(h)$. The agreement between experiment and theory confirms the validity of identifying the limit of superheating with the vortex spinodal. Another prediction of the LR theory is that for a system of particles with repulsive interactions there is no limit of supercooling. This is consistent with the data presented here.

We note that the theoretical melting line does not coincide with one of the characteristic peak effect features. This allows us to address a long-standing question regarding the peak effect, i.e., whether it is the onset, the peak, or neither [4,12–14] that signals the phase transition. Clearly, the LR melting line coincides with neither. In the two-phase coexistence model, this is not surprising because T_{on} marks the point where the disordered phase percolates while T_p is determined by the competition between the nucleation of disordered states that tend to increase the critical current and the approach to T_c, which tends to lower it.

The schematic free-energy diagrams in Fig. 4 summarize the results presented here in terms of a two-phase model. Below the spinodal line, the free-energy has two minima corresponding to disordered and ordered phases. Below the melting line $t < t_m$, the ordered phase is stable and the disordered one metastable, while for $t_m < t < t_s$ the roles are reversed. At the spinodal line, $t = t_s$, the minimum corresponding to the ordered phase is replaced by an inflection point and for $t > t_s$ the ordered phase no longer can exist.

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[1] P. M. Chaikin and T. C. Lubensky, *Principles of Condensed Matter Physics* (Cambridge University Press, Cambridge, England, 1995), 1st ed., Vol. 1.
[2] W. Henderson, E. Y. Andrei, M. J. Higgins, and S. Bhattacharya, Phys. Rev. Lett. **77**, 2077 (1996).
[3] B. Sas *et al.*, Phys. Rev. B **61**, 9118 (2000).
[4] X. S. Ling *et al.*, Phys. Rev. Lett. **86**, 712 (2001).
[5] W. Henderson, E. Y. Andrei, and M. J. Higgins, Phys. Rev. Lett. **81**, 2352 (1998); E. Y. Andrei *et al.*, J. Phys. IV (France) **10**, 5 (1999).
[6] S. N. Gordeev *et al.*, Nature (London) **385**, 324 (1997).
[7] W. K. Kwok *et al.*, Physica (Amsterdam) **293C**, 111 (1997).
[8] Z. L. Xiao, E. Y. Andrei, and M. J. Higgins, Phys. Rev. Lett. **83**, 1664 (1999).
[9] C. J. Olson *et al.*, Phys. Rev. B **67**, 184523 (2003).
[10] S. H. Autler, E. S. Rosenblum, and K. H. Gooen, Phys. Rev. Lett. **9**, 480 (1962); R. Wordenweber, P. H. Kes, and C. C. Tsuei, Phys. Rev. B **33**, 3172 (1986); S. Bhattacharya and M. J. Higgins, Physica (Amsterdam) **257C**, 232 (1996).
[11] A. I. Larkin and Yu. N. Ovchinnikov, Sov. Phys. JETP **38**, 854 (1974).
[12] W. K. Kwok, J. Fendrich, A. C. J. van der Beek, and G. W. Crabtree, Phys. Rev. Lett. **73**, 2614 (1994).
[13] P. L. Gammel *et al.*, Phys. Rev. Lett. **80**, 833 (1998).
[14] G. P. Mikitik and E. H. Brandt, Phys. Rev. B **64**, 184514 (2001).
[15] T. Giamarchi and P. Le Doussal, Phys. Rev. B **52**, 1242 (1995).
[16] E. Zeldov *et al.*, Nature (London) **375**, 373 (1995).
[17] A. Schilling *et al.*, Nature (London) **382**, 791 (1996).
[18] Y. Paltiel *et al.*, Nature (London) **403**, 398 (2000).
[19] Y. Paltiel *et al.*, Phys. Rev. B **58**, R14 763 (1998).
[20] Z. L. Xiao *et al.*, Phys. Rev. B **65**, 094511 (2002).
[21] A. E. Koshelev and V. M. Vinokur, Phys. Rev. Lett. **73**, 3580 (1994).
[22] J. Bardeen and M. J. Stephen, Phys. Rev. B **140**, A1197 (1965).
[23] M. Marchevsky, M. J. Higgins, and S. Bhattacharya, Nature (London) **409**, 591 (2001).
[24] A. Soibel *et al.*, Nature (London) **406**, 282 (2000).
[25] C. P. Bean, Rev. Mod. Phys. **36**, 31 (1964).
[26] D. W. Youngner and R. A. Klemm, Phys. Rev. B **21**, 3890 (1980); D. E. Prober, R. E. Schwall, and M. R. Beasley, Phys. Rev. B **21**, 2717 (1980). The values of H_{c2} and ξ were obtained using methods outlined by these authors to extrapolate the measured $H_{c2}(T)$ to $T = 0$. $H_{c2} = 4.21$, 4.77, and 4.84 T for samples A, B, and C, respectively.
[27] D. Li and B. Rosenstein, Phys. Rev. B **65**, 220504 (2002); cond-mat/0305258.
[28] J. E. Sonier *et al.*, Phys. Rev. Lett. **79**, 1742 (1997). The value $\lambda = 135$ nm is a two-fluid model extrapolation of the μSR measurements to $T = 0$.