Neutron Diffraction Studies of Flowing and Pinned Magnetic Flux Lattices in $2H$-$NbSe_2$

U. Yaron, P. L. Gammel, D. A. Huse, R. N. Kleiman, C. S. Oglesby, E. Bucher, B. Batlogg, and D. J. Bishop
AT&T Bell Laboratories, Murray Hill, New Jersey 07974

K. Mortensen and K. Clausen
Risø National Laboratory, 4000 Roskilde, Denmark

C. A. Bolle and F. De La Cruz
Centro Atomico Bariloche, Rio Negro, Argentina
(Received 13 June 1994)

We report on detailed neutron diffraction studies of both pinned and moving magnetic flux line lattices in $2H$-$NbSe_2$. For field-cooled, pinned lattices, we are able to make a detailed quantitative comparison with the theory of Larkin and Ovchinnikov and find excellent agreement. In studies of the dynamics, we find that the longitudinal correlation length of the vortex lattice increases substantially when it is depinned and moving due to a current exceeding the critical current, and remains large when the lattice is repinned after turning the current off.

PACS numbers: 74.60.Ec, 61.12.Ex, 74.60.Ge, 74.70.Ad

Pinning of the magnetic flux line lattice (FLL) is required in order to achieve large critical currents in a type II superconductor. Previous studies of how these lattices are pinned have tended to fall into two broad categories. There have been many studies of the structure of the FLL using techniques such as magnetic decoration [1], the scanning Hall probe [2], small angle neutron scattering [3], scanning tunneling microscopy imaging [4], and Lorentz microscopy [5]. Most of these studies tell us about the structure of the lattice when it is static and already pinned but very little about the dynamics of how it pins and unpins and what its structure is as it flows in response to an applied current. The other broad class consists of studies that probe the dynamics by measuring transport or magnetic properties such as the critical current [6,7], resistivity [8], magnetic relaxation, or susceptibility [9]. These tell us about the dynamics but require modeling and inference to extract any conclusions about the structure of the flux lattices themselves as they pin, unpin, and flow.

In this paper, we report the first detailed and quantitative study of the structure of a FLL as it is unpins and flows in response to an applied current. We have used neutron diffraction to image the lattice in a sample of $2H$-$NbSe_2$ with both current and voltage contacts so that we can systematically study the structure of the lattice as it evolves as a function of both field and applied currents. We report the observation of a number of novel effects. We see that when the system is field cooled (FC) with zero current, the resulting pinned lattice is disordered on short length scales. The evolution of this order can be followed as a function of applied field and a detailed quantitative, parameter-free comparison can be made to the theory of Larkin and Ovchinnikov (LO) [10]. The agreement is excellent. We find that this disordered FLL crystallizes into a lattice with long correlation lengths as it unpins and flows in response to an applied current. We also find that quenching the FLL from this well-ordered flowing state produces a much more ordered lattice than field cooling it from above T_c in zero current. Thus, at zero current, we can produce various different states of the flux lattice with different correlation lengths, depending on the previous history of the sample's temperature, current, and field.

Previous studies [11–14], while pioneering for their time, have provided only incomplete information on the various correlation lengths. These studies were qualitative in nature and have not analyzed the data within the framework of the concept of correlated volumes. In addition, none of these measurements studied the field and the current dependence in a systematic way, as we have in the present study.

The present experiments were performed on $2H$-$NbSe_2$. We report measurements on two single crystals whose growth are described elsewhere [15]. Sample 1 had dimensions of $6.5 \times 4.3 \times 1.0$ mm^3, and sample 2 had dimensions of $4.8 \times 4.6 \times 2.0$ mm^3. Both x-ray and neutron diffraction studies confirmed the $2H$ polytype and determined the **c** axis mosaic to be less than $0.1°$ FWHM. Magnetization and transport studies of both crystals showed sharp superconducting transitions, less than 40 mK wide at all fields. For sample 1 $T_c(0) = 7.2$ K and for sample 2 $T_c(0) = 7.4$ K.

We report here on samples studied using the small angle neutron scattering spectrometer in the cold neutron guide hall of the Risø DR3 reactor. In the experiment a superconducting magnet in the persistent mode was used to apply a horizontal field parallel within $1°$ to both the **c** axis of the crystal and the neutron beam. The samples were masked with Cd foil to expose a $\sim 4 \times 4$ mm^2 region to the neutron beam, leaving all electrical contacts masked to reduce the small angle scattering background.

We used an incident neutron wavelength of $\lambda_n \sim 5-18$ Å and a bandwidth of $\Delta\lambda_n/\lambda_n \sim 18\%$ or 36%, with an incident beam divergence of $\sim 0.18°$ and $\sim 0.1°$ FWHM in the horizontal and vertical directions. The diffracted neutrons were counted by an area detector located at the end of a 6 m evacuated chamber. The scattering geometry allows us to study all three correlation lengths. The two positional correlation lengths perpendicular to the flux lines $\xi_p^{r\parallel G}$ and $\xi_p^{r\perp G}$, which roughly correspond to compressional and shear displacements of the lattice, respectively, can be extracted from the radial and azimuthal widths of the six first order scattering peaks on the Ewald sphere, each of which can be brought down onto the plane of the detector with appropriate rotating and tilting of the cryostat. The longitudinal correlation length ξ_L which is a measure of correlations parallel to the direction of the flux lines is extracted from the widths of the rocking curves.

We have studied the field dependence and the effect of different magnetic and current histories [field cooled (FC), zero field cooled (ZFC), and high field cooled (HFC), a process in which the sample is cooled with $H > H_{c2}$] on both the locations and widths of the six lowest order peaks. The locations of the peaks indicate that the flux lattice always remains aligned to the crystalline **a** axis for all fields, histories, and currents studied. At different fields, the peaks are always found to occur, to within the measurement errors, at $\tau = 2\pi(2B/\sqrt{3}\phi_0)^{1/2}$, where τ is the scattering vector. To within the resolution, we have not found any evidence of asymmetries in the intensities, widths, or locations of the six lowest order peaks as a function of current or field history. All three procedures gave rise to resolution limited peaks in the azimuthal and radial directions for all fields. This implies long ranged orientational order and a lower limit on $\xi_p^{r\perp G}$ and $\xi_p^{r\parallel G}$ of roughly 0.2 μm at 500 Oe. On the other hand, the widths of the rocking curves were sensitive to the process used to form the FL. At $H = 2$ kOe the ZFC and HFC processes produced ξ_L of ~ 5 and >12 μm, respectively. At this field, the FC process produced peaks with an intermediate value of $\xi_L \sim 8$ μm. For fields $H/H_{c2} < 0.15$ ($H_{c2} = 22$ kOe), increasing the field caused a narrowing of the peak widths indicative of an increase of ξ_L, which becomes resolution limited for fields above this. Controlling the cooling rate between ~ 0.5 K/min and ~ 1 K/sec did not alter the observations discussed above. We believe that the fact that a HFC procedure generates lattices with a longer longitudinal correlation length can be understood as arising from the fact that the more well correlated the initial state, the better the final lattice. At high fields, interactions dominate and one pins a more ordered structure than from a ZFC lattice where pinning dominates and interactions are relatively less important. The low critical currents of our samples ($J_c \sim 30-50$ A/cm^2) are crucial to seeing this response as a large J_c would produce field gradients in a HFC or ZFC experiment which would obscure these effects.

Shown in Fig. 1 are the FWHM rocking curve widths σ_m for sample 2 as a function of field at a temperature of 4.7 K for FC lattices. These widths were obtained from two different types of analyses. In the first, the widths were obtained from the directly measured rocking curves and calculated using $\sigma_m^2 = \sigma_{\text{rock}}^2 - \sigma_{\text{res}}^2$, where σ_{res} is the instrumental resolution. In the second type of analysis, they were extracted from the radial widths of the diffracted spots on the plane of the detector itself at the peak of the rocking curves using the relation [16] $(\text{FWHM}_{\text{det-rad}})^2 = [a^2(b^2 + c^2) + (2bc)^2]/(a^2 + b^2 + c^2) + d^2$, where a is the beam divergence, $b = \sigma_m$, $c = \theta\Delta\lambda_n/\lambda_n$ is the energy divergence, and d is the detector smearing. Both techniques give the same values for σ_m, implying that we have a good understanding of the resolution function. The possibility to obtain σ_m by these two ways results from the fact that in reciprocal space the radial direction on the plane of the detector is not exactly orthogonal to the rocking curve. One should not confuse the radial direction on the plane of the detector with the pure radial θ-2θ scan, which is directly related to the compressional order of the lattice, as discussed earlier. Extracting σ_m from the widths of the peaks on the detector has the advantage that it requires less counting time, and we will exploit this in our experiments on the effects of current to be discussed later.

Shown in Fig. 2 are the longitudinal correlation lengths as a function of magnetic field extracted from the FC

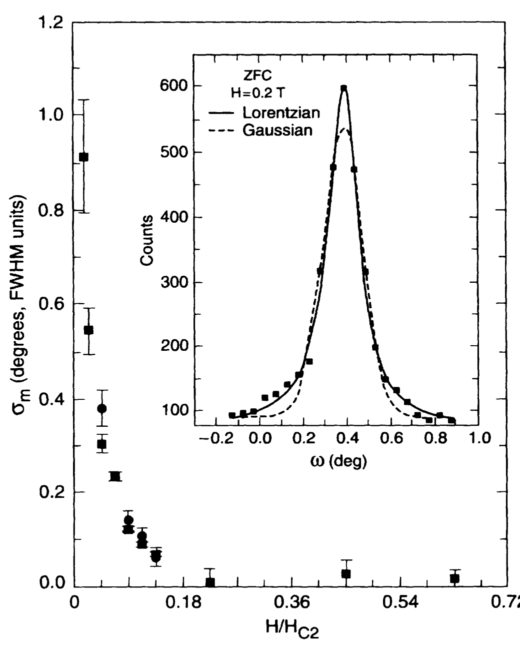

FIG. 1. The field dependence of σ_m taken at 4.7 K, extracted from full rocking curves (squares) and from an analysis of the radial width of the spot on the plane of the detector at the peak of the rocking curve (circles). The inset shows a rocking curve measured at 2 kOe following a ZFC process, fitted by a Gaussian (dashed) and Lorentzian (solid).

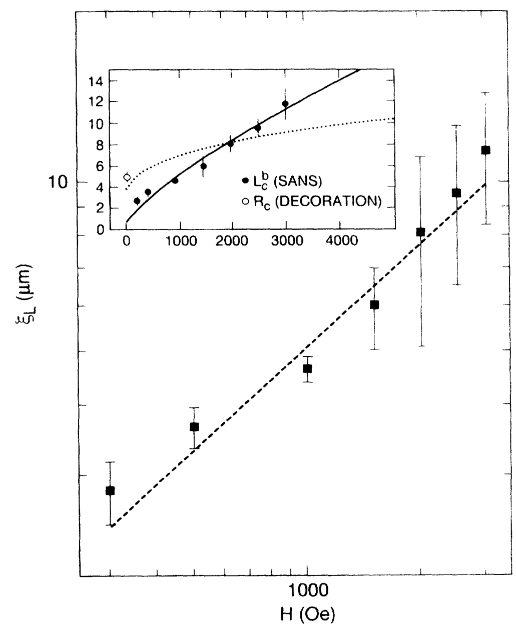

FIG. 2. The field dependence of ξ_L on a log-log plot at 4.7 K. The inset shows these values (filled circles) on a linear plot with the prediction of the collective pinning theory of Larkin and Ovchinnikov (solid line). The dotted line is the prediction for R_c and the open circle is the result of a decoration experiment.

data in Fig. 1, using the relation $\xi_L = 1/\Delta\tau = 1/\tau\sigma_m = (\frac{1}{2}\pi)[(3)^{1/2}\phi_0/2B\sigma_m^2]^{1/2}$. Our data show that, over the field range in which we could follow it, ξ_L increases monotonically with field with a nonzero intercept at zero field. By independently measuring the critical current for this sample we can use our neutron data to perform the first quantitative test [17] of the predictions of the LO theory. In that theory, the longitudinal correlation length of a bundle of vortices L_c^b (or ξ_L in our notation) in the appropriate limit should be given by $\sim \lambda R_c/a_0$, where R_c is a positional correlation length given by $\xi(J_0/J_c)^{1/2}$, where J_0 is the depairing critical current, and λ is the superconducting magnetic penetration depth. This simplifies to $L_c^b = A(\xi c\phi_0/a_0^2 J_c 6\sqrt{12}\pi^2)^{1/2}$, where A is a prefactor of order unity, ξ is the superconducting coherence length, and c is the speed of light. Shown in the inset of Fig. 2 as the solid line is the prediction for L_c^b of the theory using our measured critical currents. The measured values of L_c^b are the filled circles. Several points should be noted. The magnitudes of L_c^b predicted from theory agree well with the data, with the prefactor A for the line shown being 0.5. The fit of the theory to the measured field dependence of L_c^b is also excellent. This represents the first direct quantitative test of this theoretical prediction, and we find agreement (to within a factor of 2) for both the absolute magnitude as well as the field dependence of L_c^b. We can also compare the theory with the measured value of R_c as obtained from a decoration experiment. Shown as the dotted line in the inset to Fig. 2 is the quantity $a_0 L_c^b/\lambda$. The open circle is the value obtained from a magnetic decoration experiment, as described elsewhere [1]. The theory fits both experiments quite well. Finally, the theory also makes a prediction about the line shapes: The rocking curves should be better fit by Lorentzians than Gaussians. This can be seen simply as the fact that the Fourier transform of an exponential is a Lorentzian. The better fit of the Lorentzian to our data is obvious (inset to Fig. 1).

Shown in Fig. 3 are the effects on the radial width of the spot on the plane of the detector at the peak of the rocking curve of a transverse transport current applied in a direction perpendicular to the external field. The data were taken on sample 1 at 4.7 K, which was field cooled at $H = 2.5$ kOe with no current applied while cooling. After cooling, the measurements were performed upon increasing the current from zero to $2L_c$ ($L_c \sim 1$ A, $J_c \sim 50$ A/cm^2) and then decreasing it back to zero. The different symbols refer to different current sequences as described in the figure. Each experimental point corresponds to roughly 15 min of counting. After field cooling with zero current, we find a flux crystal with a σ_m of 0.19°, which corresponds to a longitudinal correlation length of $\xi_L \sim 40a_0$. As is evident from the figure,

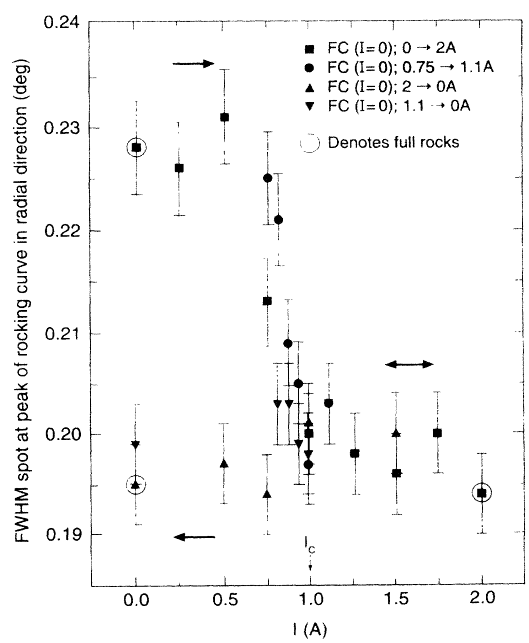

FIG. 3. FWHM of the radial width of the spot on the plane of the detector at the peak of the rocking curve as a function of the applied current following a FC process at 2.5 kOe and 4.7 K. Each data point corresponds to a measurement of ~15 min. Data shown here were collected using four different current sequences as described in the figure. The circled data points were obtained from full rocking curves.

applying a transport current which is significantly smaller than the critical current does not, to within our resolution, have any effect on ξ_L. However, when $L \sim L_c$, we find an abrupt annealing of the lattice as evidenced by the FWHM dropping, corresponding to $1/\xi_L < 1/80a_0$ for our flux crystal. For currents above L_c, we continue to see well ordered flux crystals. The well ordered flux crystal remains after the transport current is decreased back to zero. This is independent of the rate at which this current is removed. Thus a cycling of the applied current allows us to anneal away the effects of pinning disorder as evidenced by a measurement of ξ_L in the flux crystal. Full rocking curves taken as marked on Fig. 3 quantitatively agree with this picture. A complete set of rocking curves collected on sample 2 using a conventional spectrometer (to be reported elsewhere) are also consistent with the results described above.

Our observation of flux lattice states with varying degrees of disorder all at the same temperature, field, and zero current implies that under these conditions the lattice does not rearrange itself on length scales longer than a minimum pinning length on the time scales (minutes to hours) of this experiment. When we field cool in zero current, the disordered state is produced for one of at least two possible reasons: Either (i) this is the equilibrium state and the cooling rates are sufficiently slow that the system can find it or (ii) the system rapidly enters a disordered, pinned nonequilibrium state as it passes through H_{c2} and remains disordered as it is further cooled because the pinning free energy barriers are too high to cross. When the applied current is increased above L_c, the lattice flows past the pinning potential, averaging out its effects so the vortex-vortex interactions become more dominant and induce a longer correlation length [18]. When the current is then turned off, there are again two possibilities corresponding to the above scenarios: Either (i) the system is stuck by the high pinning barriers in the more well ordered state even though a more disordered state has a lower free energy or (ii) the more ordered state actually has a lower free energy so the system simply remains there.

In conclusion, we have presented the results of a quantitative study of the correlations of a magnetic flux line lattice as a function of field and with applied currents below, at, and above the critical current. For field-cooled, pinned lattices we have quantitatively tested the LO theory and find it to work quite well. We also report the observation of a number of novel effects for flux lattices as they are unpinned. Field-cooled lattices, which at low fields have only short ranged correlations, are found to crystallize into well-ordered lattices when they unpin and flow. Quenching lattices from the flowing state is found to produce much more well-ordered structures than are formed if one field cools with zero applied current.

The authors would like to acknowledge many helpful discussions with G. Aeppli, C. Broholm, S. N. Coppersmith, D. S. Fisher, D. R. Nelson, and H. Safar. We would like to thank H. L. Williams for x-ray data on both crystals. One of us (U. Y.) would like to acknowledge the Lady Davis Fellowship Trust and the Israeli Ministry of Science and the Arts for financial support.

[1] C. A. Bolle *et al.*, Phys. Rev. Lett. **71**, 4039 (1993).
[2] A. M. Chang *et al.*, Appl. Phys. Lett. **61**, 1974 (1992).
[3] E. M. Forgan *et al.*, Nature (London) **343**, 735 (1990); B. Keimer *et al.*, Science **262**, 83 (1993); P. L. Gammel *et al.*, Phys. Rev. Lett. **72**, 278 (1994).
[4] H. F. Hess *et al.*, Phys. Rev. Lett. **69**, 2138 (1992).
[5] K. Harada *et al.*, Phys. Rev. Lett. **71**, 3371 (1993).
[6] P. Koorevar *et al.*, Phys. Rev. B **42**, 1004 (1990).
[7] S. Bhattacharya and M. J. Higgins, Phys. Rev. Lett. **70**, 2617 (1993).
[8] T. W. Ting and N. P. Ong, Phys. Rev. B **42**, 10781 (1990).
[9] C. A. Duran *et al.*, Phys. Rev. B **44**, 7737 (1991).
[10] A. I. Larkin and Yu. Ovchinnikov, J. Low Temp. Phys. **34**, 409 (1979); G. Blatter *et al.* (to be published).
[11] D. Cribier *et al.*, Phys. Rev. Lett. **28**, 1370 (1972).
[12] Y. Simon and P. Thorel, Phys. Lett. **35A**, 450 (1971).
[13] J. Schelten *et al.*, Phys. Rev. B **12**, 1772 (1975).
[14] P. Thorel *et al.*, J. Phys. (Paris) **34**, 447 (1973).
[15] C. S. Oglesby *et al.*, J. Cryst. Growth **137**, 289 (1994).
[16] R. N. Kleiman, thesis, Cornell University, 1992 (unpublished); R. Cubitt *et al.*, Physica (Amsterdam) **180–181B**, 377 (1992).
[17] A qualitative comparison has been made by D. K. Christen *et al.*, IEEE Trans. Magn. **19**, 884 (1983).
[18] A. Schmid and W. Hauger, J. Low. Temp. Phys. **11**, 667 (1973).