Observation of Tilt Induced Orientational Order in the Magnetic Flux Lattice in $2H$-$NbSe_2$

C. A. Bolle and F. De La Cruz
Centro Atomico Bariloche, Rio Negro, Argentina

P. L. Gammel, J. V. Waszczak, and D. J. Bishop
AT&T Bell Laboratories, Murray Hill, New Jersey 07974
(Received 10 June 1993)

We report on observations of the magnetic flux lattice in $2H$-$NbSe_2$ using the magnetic decoration technique. For low applied magnetic fields and with the field applied parallel to the c axis of the crystal, we find a disordered flux lattice with short-ranged positional and orientational order. As the tilt angle of the applied magnetic field increases, we find that the orientational order in the flux lattice grows dramatically. At high tilt angles, we find a hexatic solid with long-ranged orientational order but only short-ranged positional order. This is the first observation of a flux lattice in which the orientational order is stabilized by changing the tilt angle of the applied magnetic field in an anisotropic superconductor.

PACS numbers: 74.60.Ge

The high-T_c superconductors have challenged the conventional wisdom and in some cases forced us to change our understanding of both the statics and dynamics of flux lattices in anisotropic systems [1]. In particular, a number of novel static structures have been observed, some of which were predicted but most of which were not. These include orientationally ordered hexatic flux lattices [2], vortex chains of three different types [3–5], sawtooth vortex lattices [6], order-disorder transitions [7], square flux lattices [8], oval vortices [9], distorted hexagonal lattices [9], intrinsic pinning [10], and pinning to twin boundaries [10]. It is clear that these static structures provide a stringent test of our present theories. In addition, these static structures provide the essential starting point for the interpretation of many experiments which study the dynamics of vortices in these materials such as NMR, μSR, and neutron scattering line shapes.

An important issue in the study of the flux lattices in these materials is the type of long-range order present. Previous studies [2,7,11] have shown that at high fields, the lattices in BiSrCaCuO (BSCCO) and YBaCuO (YBCO) are hexatics with long-ranged orientational order but with only short-ranged positional order. At low fields, the lattices in these materials are quite disordered and look liquidlike with only short-ranged, exponentially decaying positional and orientational order. In YBCO and BSCCO this transition to a hexatic phase at high fields is an abrupt one, driven by increasing the magnetic field [7]. In this paper we show that orientational order can also be stabilized in an anisotropic superconductor by tilting the applied magnetic field away from the c axis [12]. Thus, at high magnetic fields and/or large tilting angles, one finds that the flux lattices in an anisotropic superconductor are hexatic while at low fields and for small angles they are liquidlike. The effect we report here is similar to that seen in smectic liquid crystals [13] where it is found that a spontaneous tilt of the liquid crystal molecules can stabilize long-ranged orientational order.

Our experiments were performed on the conventional but moderately anisotropic superconductor $2H$-$NbSe_2$. This material has an in-plane penetration depth of 2000 Å and a mass anisotropy $\Gamma=\gamma^2=m_c/m_a\sim 11$. This compares with a Γ of 60 for YBCO [9,14], 3000 or more for BSCCO [15], and 100 to 400 for LaSrCuO [16]. The crystal structure of this material is hexagonal and layered, making it a good candidate to study the effects of anisotropy on flux lattice structures in a moderately anisotropic system. Our samples were typically hexagonally shaped slabs which were 2–3 mm on a side and $\sim$200 μm thick with a sharp resistive transition at 7.0 K. We imaged the flux lattices in these samples using high resolution Bitter patterns. The technique has been described in detail elsewhere [17].

In our measurements, the crystals were glued to the sample holder with silver paint and a thin layer of material was cleaved off of the surface just prior to cooling the sample for decoration of the flux lattice with iron smoke. This allowed us to use a single crystal for more than one applied field. We have used magnetic fields in the range 10–70 Oe applied at angles from 0° to 70° with respect to the c axis. After decoration, the sample was warmed up and the lattice imaged in an SEM. Typically we took 24, nonoverlapping images from different parts of the same crystal with 800–1000 vortices in each image. These pictures were digitized for the analysis which we will describe below. This analysis of the patterns was then for an averaged response over distances of several hundred microns across the surface of the samples.

Typically, each image had 512×400 pixels with a dynamic range of 4096 gray levels. The vortices were located in such images by intensity maximization of a 3×3 pixel sliding window. Our subsequent analysis determined the nearest neighbors, the Fourier transform of the set of points, and calculated the orientational and positional correlation functions. Three reciprocal lattice vectors for the positional correlations were extracted from the peaks of the Fourier transform of each image and from these we calculated 72 positional and 24 orientational correlation functions for each sample. The results

presented here are an average of these functions.

We will start the presentation of our results by discussing the data for the magnetic field applied parallel to the c axis. The flux lattices we find for this field orientation resemble a polycrystal with the typical size of each crystallite being several lattice constants in diameter. We find that for this orientation the defects tend to form grain boundaries and not to be single and isolated. This is very different from the behavior found in the high-T_c materials YBCO and BSCCO in which there is a uniform concentration of isolated defects producing the short-ranged order but no grain boundaries in the flux lattice. Figure 1(a) shows the evolution of the angular (ellipse) averaged orientational correlation function $G_6(r)$ as a function of increasing field applied parallel to the c axis. It can clearly be seen that increasing the field has only a small effect on the orientational order of the flux lattice. No sharp order-disorder transition is seen as was observed for BSCCO and YBCO as a function of applied field. The correlation length for this field orientation is

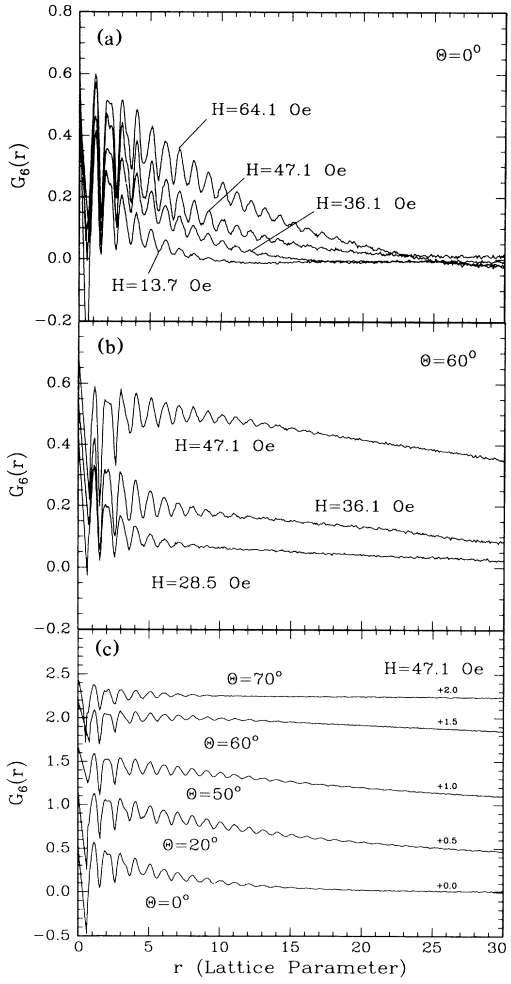

FIG. 1. (a) Field dependence of the orientational correlation function $G_6(r)$ for $\theta=0°$ and four different applied fields. (b) Same dependence for $\theta=60°$ and three different applied fields. (c) Angular dependence of $G_6(r)$ at a constant applied field of 47.1 Oe and five different tilting angles θ.

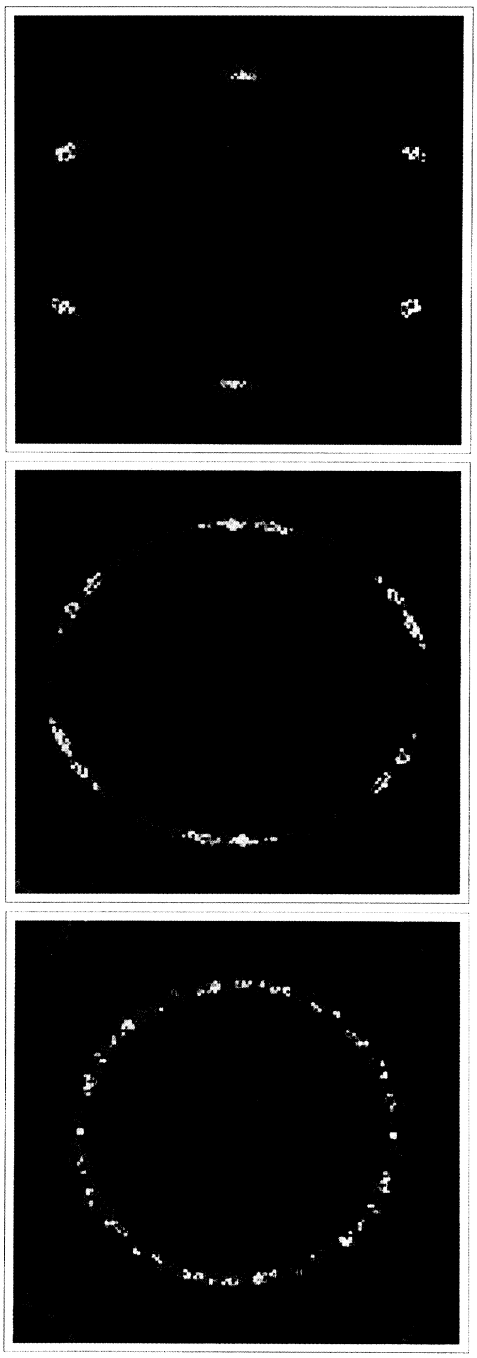

FIG. 2. Angular dependence of the Fourier transform of the flux line lattice for 47.1 Oe applied field and $\theta=70°$ (top), $\theta=50°$ (center), and $\theta=30°$ (bottom) tilting angle. The sample rotation axis is parallel to the short edge of the picture.

always less than 10 lattice constants.

Another feature which evolves very gradually in our data is the locking-in of the vortex structure to the crystal structure as the field is changed. As we increase the field, we see flux crystals with orientations close to the crystalline a axis lock into this orientation. This produces Fourier transforms which show an isotropic liquidlike ring of scattering at low fields which evolves into a ring structure with six well-defined peaks at higher fields.

Shown in Fig. 2 is the central result of this paper. We have plotted the Fourier transform of the flux lattices at a fixed field of 47.1 Oe for three angles of the field with respect to the c axis. In the bottom part of the figure $\theta=30°$, in the center part $\theta=50°$, and in the upper part $\theta=70°$. One clearly sees two effects. As the tilt angle increases, the ring is foreshortened and becomes more elliptical for increasing tilt angle. At the same time, the isotropic ring of scattering evolves into a sixfold modulation and at the highest angle into fully resolved spots. These data show a clear locking-in to the rotation axis at high angles from a disordered lattice at low angles. Increasing the tilt angle clearly stabilizes the orientational order of the flux lattice in this system. We note that the flux lattice orientation is uncorrelated with the crystal lattice so this result is *not* induced by the substrate. This is an effect similar to that seen in smectic liquid crystals [13] where the tilted phases are found to be hexatics with very long-ranged orientational order. In the liquid crystal case, the tilting is a spontaneous effect due to the structure of the molecules themselves while here the tilting is obviously externally induced by the rotation of the magnetic field angle, but the results for the underlying lattices and their orientational order are the same.

The qualitative behavior seen in the Fourier transforms of Fig. 2 can be made more quantitative. Shown in Fig. 1(b) is the decay of the orientational order at a fixed angle of 60° for a number of applied fields. At this fixed angle both the magnitude of the orientational correlations and their range increase dramatically with increasing field. While for $\theta=0°$ the decay lengths are found to be in the range of 4–7 lattice constants, for similar fields at $\theta=60°$ the decay lengths are found to vary between 22 and 55 lattice constants. In Fig. 1(c) we show the orientational correlations at a fixed field for various tilt angles. As the tilt angle increases, we see a clear evolution from short-ranged orientational order for small angles to a hexatic phase with long-ranged orientational order at large angles. For the data shown in Fig. 1(c), the correlation lengths change from 7 to 350 lattice constants. A closer examination of the short-ranged behavior of the correlation functions as a function of angle is instructive. $G_6(r=a_0)$ starts out at 0.57 for $\theta=0°$, it reaches a maximum of 0.63 at $\theta=20°$, and then falls to 0.38 at $\theta=70°$. This suggests that at $\theta=70°$ we have a state with significant dispersion in the bond angles but the overall orientation parallel to the rotation axis is kept for distances as long as 350 lattice constants.

The functional form for the orientational correlation functions is found to be most consistent with stretched exponentials for $\theta<50°$. In this range the data fit well to the form $G_6(r)\sim\exp[-(r/\Lambda)^{\zeta}]$ with ζ in the range $1.0<\zeta<1.7$ with no clear trend being apparent for this exponent. For angles above 50° the best fits are found for pure exponentials or $\zeta=1$.

In Fig. 3 we show a plot of the fitted orientational correlation lengths Λ as a function of both the tilting angle and the lattice parameter $a_0=(2\phi_0/\sqrt{3}H)^{1/2}$. One can clearly see the gradual increase of Λ with increasing field (or decreasing a_0) and the dramatic increase with increasing tilt angle. This diverging correlation length with increasing tilt angle is the signature of the tilt angle induced orientational order in this system. We find a very different behavior for the positional order. In the entire angle-field range we have studied, we find that the best fits to the positional correlation functions are stretched exponentials with $G_g(r)\sim\exp[-(r/\rho)^{\xi}]$. Typical values for ξ lie in the range 0.2–0.6. There is no clear variation of ρ with field or angle, the values always being around one lattice constant. The positional order in this system is then always very short ranged over the entire field-angle plane we have studied. At high angles, the system has only orientational order but not positional order. It is a hexatic.

We can compare the orientationally ordered lattice we find at high fields and large tilt angles with the predictions of either uniaxial London or Ginzburg-Landau theory [12]. These models, which calculate the bulk interaction for vortex lines and ignore surface terms, would predict for a mass ratio of 10, a set of flux chains running perpendicular to the rotation axis as has been seen in YBCO [4]. Our observation of orientational order as described in this paper is the precursor to chains running at

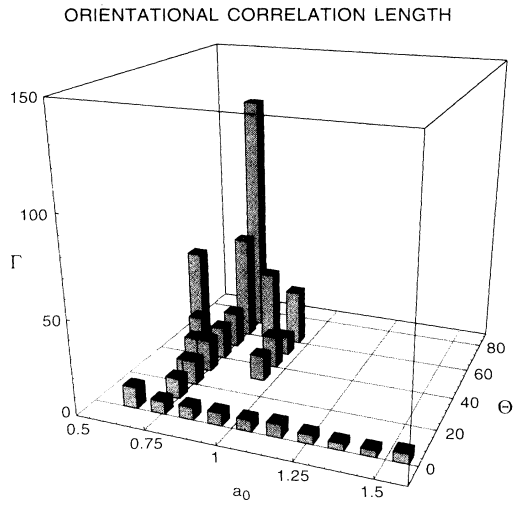

FIG. 3. Orientational correlation length Λ as a function of the lattice parameter $a_0=(2\phi_0/\sqrt{3}H)^{1/2}$ and the tilting angle θ.

90° to this direction as has been seen in scanning tunneling microscopy studies [5]. In the theoretical models with only bulk interactions, this is the higher energy direction. These models should apply for moderate anisotropy materials in the London limit. This applies to YBCO and in that material one finds very good agreement with bulk theory. BSCCO is very anisotropic and while one finds vortex chains in that system [3], they disagree with theory in that one finds both chain vortices as well as Abrikosov vortices between the chains. $2H$-$NbSe_2$ is a weakly anisotropic material for which the bulk theories should apply but they do not and we speculate that this could be due to surface terms. On the basis of calculations done in Ref. [18], one can show [19] that for angles θ less than $\tan^{-1}\gamma$ ($\sim 70°$ for this material) the surface terms favor the orientation that we see in the experiment whereas the bulk terms in the free energy expansions favor an orientation at right angles to what we see. The contributions to the free energy from the surfaces should occur for distances of order a penetration depth from the surface. Because the sample is much thicker than the low temperature penetration depth, one might expect that these terms would be unimportant. We suggest that they may be much more important in $2H$-$NbSe_2$ than in YBCO because in YBCO the vortex patterns are frozen in well below the mean field H_{c2} and the penetration depth is then approaching its low-temperature value producing a situation in which the bulk terms dominate. However, in $2H$-$NbSe_2$ the vortex patterns should be frozen in very close to the mean field H_{c2} because of the much weaker effects of thermal fluctuations in this system. The resulting longer penetration depth might make the surface terms much more important. To be more quantitative, in the London approximation, the energy differences δF between different flux lattices as estimated in Ref. [12] for bulk interactions are very small: $\delta F/F \sim 10^{-3}\phi_0/B\lambda^2(T)$ so increasing $\lambda(T)$ dramatically lowers the free energy advantage for the favored orientation, opening the way for other terms, such as surface interactions, to dominate the patterns. The weaker thermal fluctuations and the increased importance of the surface terms in this material may allow the surfaces to play an important role in producing the flux patterns that we see.

In conclusion, we have presented data on the magnetic flux lattice in $2H$-$NbSe_2$ as imaged using magnetic decoration. We have shown that increasing the tilt angle in this system can induce orientational order. At high tilt angles we find a flux lattice which is a hexatic with long-ranged orientational order but only short-ranged positional correlations. Present theories in the London limit are inconsistent with our results presumably because they ignore the surface terms. The issue of the discrepancy between the observed and predicted static vortex structures in anisotropic superconductors is an important one as it relates to the basic vortex-vortex interaction. Clearly more theoretical and experimental work is needed.

The authors would like to thank David Huse, John Clem, Ron Pindak, Cherry Murray, Harald Hess, Carlos Durán, David Nelson, and Vladimir Kogan for many helpful discussions. We would like to acknowledge S. Dutrus and C. Cotaro for their generous assistance with the SEM.

[1] D. J. Bishop, P. L. Gammel, D. A. Huse, and C. A. Murray, Science **255**, 165 (1992).
[2] C. A. Murray, P. L. Gammel, and D. J. Bishop, Phys. Rev. Lett. **64**, 2312 (1990).
[3] C. A. Bolle, P. L. Gammel, D. G. Grier, C. A. Murray, D. J. Bishop, D. B. Mitzi, and A. Kapitulnik, Phys. Rev. Lett. **66**, 112 (1991).
[4] P. L. Gammel, D. J. Bishop, J. P. Rice, and D. M. Ginsberg, Phys. Rev. Lett. **68**, 3343 (1992).
[5] H. F. Hess, C. A. Murray, and J. V. Waszczak, Phys. Rev. Lett. **69**, 2138 (1992).
[6] P. L. Gammel, C. A. Durán, and D. J. Bishop, V. G. Kogan, M. Ledvij, and A. Yu. Simonov, J. P. Rice, and D. M. Ginsberg, Phys. Rev. Lett. **69**, 3808 (1992).
[7] D. G. Grier, C. A. Murray, C. A. Bolle, P. L. Gammel, D. J. Bishop, D. B. Mitzi, and A. Kapitulnik, Phys. Rev. Lett. **66**, 2270 (1991).
[8] M. Yethiraj, H. A. Mook, G. D. Wignall, R. Cubitt, E. M. Forgan, D. M. Paul, and T. Armstrong, Phys. Rev. Lett. **70**, 857 (1993); R. N. Kleiman *et al.*, Bull. Am. Phys. Soc. **38**, 579 (1993).
[9] G. J. Dolan, F. Holtzberg, C. Field, and T. R. Dinger, Phys. Rev. Lett. **62**, 2184 (1989).
[10] G. J. Dolan, G. V. Chandrasekar, T. R. Dinger, C. Field, and F. Holtzberg, Phys. Rev. Lett. **62**, 827 (1989).
[11] P. L. Gammel (unpublished).
[12] L. J. Campbell, M. M. Doria, and V. G. Kogan, Phys. Rev. B **38**, 2439 (1988); L. N. Bulaevskii, M. Ledvij, and V. G. Kogan, Phys. Rev. B **46**, 366 (1992).
[13] R. Bruinsma and D. R. Nelson, Phys. Rev. B **23**, 402 (1981); J. D. Brock *et al.*, Phys. Rev. Lett. **57**, 98 (1986).
[14] D. E. Farrell, C. M. Williams, S. A. Wolf, N. P. Bansal, and V. G. Kogan, Phys. Rev. Lett. **61**, 2805 (1988); U. Welp, W. K. Kwok, G. W. Crabtree, K. G. Vandervoort, and J. Z. Liu, Phys. Rev. Lett. **62**, 1908 (1989).
[15] D. E. Farrell, S. Bonham, J. Foster, Y. C. Chang, P. Z. Jiang, K. G. Vandervoort, D. J. Lam, and V. G. Kogan, Phys. Rev. Lett. **63**, 782 (1989).
[16] B. Janossy, H. Kojima, I. Tanaka, and L. Fruchter, Physica (Amsterdam) **176C**, 517 (1991).
[17] P. L. Gammel, D. J. Bishop, G. J. Dolan, J. R. Kwo, C. A. Murray, L. F. Schneemeyer, and J. V. Waszczak, Phys. Rev. Lett. **59**, 2592 (1987).
[18] V. G. Kogan, A. Yu. Simonov, and M. Ledvij, Phys. Rev. B **48**, 392 (1993).
[19] V. G. Kogan (private communication).

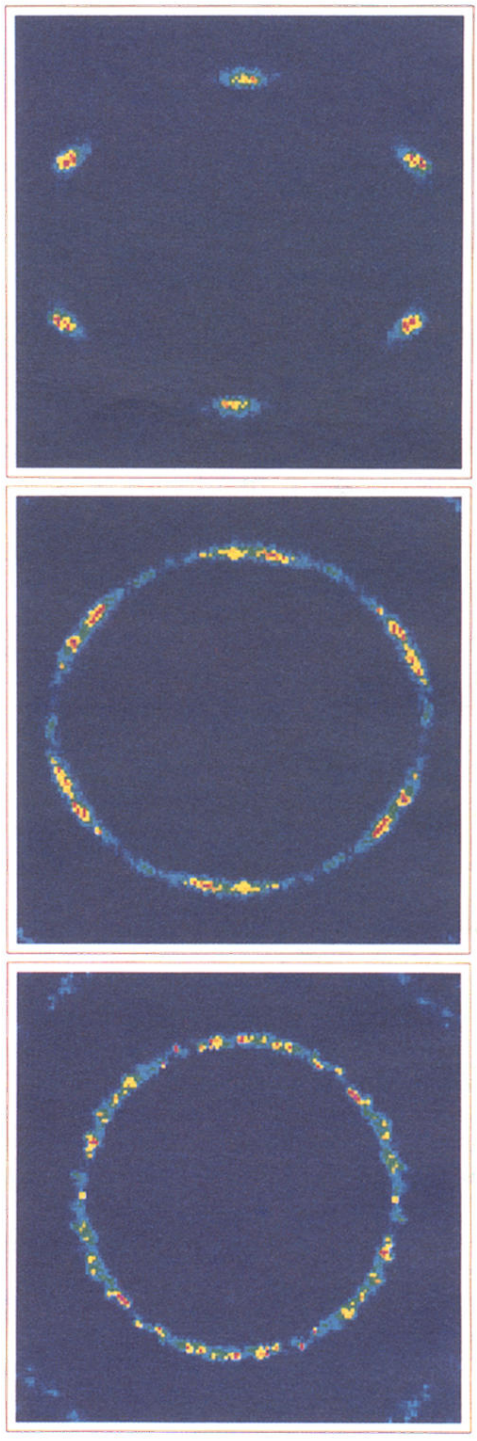

FIG. 2. Angular dependence of the Fourier transform of the flux line lattice for 47.1 Oe applied field and $\theta = 70°$ (top), $\theta = 50°$ (center), and $\theta = 30°$ (bottom) tilting angle. The sample rotation axis is parallel to the short edge of the picture.