Intrinsic Behavior of Flux Lines in Pure Niobium near the Upper Critical Field

E. M. Forgan,[1] S. J. Levett,[2,3] P. G. Kealey,[1] R. Cubitt,[3] C. D. Dewhurst,[3] and D. Fort[4]

[1]*School of Physics and Astronomy, University of Birmingham, Birmingham B15 2TT, United Kingdom*
[2]*Department of Physics, University of Warwick, Coventry CV4 7AL, United Kingdom*
[3]*Institut Laue-Langevin, 38042 Grenoble, France*
[4]*School of Metallurgy and Materials, University of Birmingham, Birmingham B15 2TT, United Kingdom*

We report small-angle neutron-scattering (SANS) measurements of flux line properties near H_{c2} in an ultrapure sample of niobium with weak pinning of flux in the bulk. These confirm in detail the Abrikosov picture of the flux line lattice to within 20 mK of the upper critical field line. However, it has recently been claimed [X. S. Ling *et al.,* Phys. Rev. Lett. **86**, 712 (2001)], on the basis of SANS observations of a disordering of flux lines in niobium, that the flux lattice melts at temperatures clearly separated from the upper critical field line. This discrepancy may possibly arise from differences in sample purity and pinning.

In recent years, there has been great and continuing interest in the properties of magnetic flux lines in superconductors. This is partly because thermal effects are much enhanced in high T_c materials. Hence, *melting* of the Abrikosov [1] flux line lattice (FLL) was both predicted [2,3] and directly observed (see, for example, Refs. [4,5]). Another strong influence on the FLL is any quenched disorder in the underlying crystal lattice, which can pin flux lines and reduce the long range order [6]. If the pinning is sufficiently strong, the FLL can be destroyed and a glassy flux line state will result. Detailed reviews of the interplay of thermal excitation, quenched disorder, and material properties on vortex matter are given, e.g., in Refs. [7,8]. One general result, first noticed by Pippard [9] and subsequently elaborated by others, is that the "softer" the FLL the more readily it is pinned. Pippard [9] invoked the softening of the shear modulus of a FLL near H_{c2} as an explanation of the "peak effect," which is a big increase in the pinning of flux lines observed just below (in temperature or field) the $H_{c2}(T)$ line.

There has also been great interest in whether high T_c ideas also apply to materials with T_c of order 10 K. In particular, there is a recent report [10] of a hysteretic first order transition in niobium: between a state giving six clear neutron diffraction spots from a hexagonal FLL, and another state closer to H_{c2} giving a liquid-like ring of diffracted intensity. It is claimed [10] that this first order transition represents the melting of the FLL into a liquid state. However, it is important to determine whether this transition is an intrinsic property of pure niobium or arises because of a destruction of FLL order associated with the peak effect, which was observed in the sample. We have therefore carried out a small-angle neutron-scattering (SANS) investigation of the FLL structure close to H_{c2} in an ultrapure sample of niobium with a very low level of bulk pinning of flux lines.

After growth, our single crystal of Nb was annealed under UHV for a week at a few hundred °C below its melting point. The residual resistance ratio is ~10 000, corresponding to an electron mean free path $\ell > 1000 \times$ the superconducting coherence length ξ. From the crystal, an oval plate (diam 7×9 mm^2, thickness 2.46 mm) was spark cut, with a [111] crystal direction perpendicular to the plane of the plate. Any surface damage due to spark cutting was removed with an HF/HNO$_3$ polishing etch. The surface was then *very gently* roughened with a fine abrasive to induce strong surface pinning. This would hold the magnetic induction essentially constant during a temperature scan at constant applied field, while allowing the FLL inside to move freely. (This technique was used in high resolution heat capacity measurements [11] on a similar sample.) The specimen was mounted inside a sapphire-windowed cryostat placed between the poles of an electromagnet applying a field of 0.2 T parallel to the [111] direction. A neutron-absorbing Cd mask with a 5-mm-diameter hole was placed over the center of the plate sample so that neutrons, which were incident approximately parallel to the field, would be diffracted only by flux lines in the bulk.

The measurements were carried out on the SANS diffractometer D22 at the ILL, used in a high-resolution mode ($\lambda = 10$ Å neutrons; collimation and detector distances ~18 m). To maximize the intensity for any particular diffraction spot, the sample and electromagnet together could be rocked in a horizontal or vertical direction to the Bragg angle for that spot. It is clear from the FLL diffraction pattern shown in Fig. 1 that a well-ordered hexagonal FLL is formed. The sample temperature was raised from 3.2 K in small increments through the $H_{c2}(T)$ line. At each temperature, we measured a rocking curve of the intensity of the horizontal "(1, 0)" spot on the right of Fig. 1, versus sample angle. Similar data were taken on cooling. The angular width of the rocking curve gives the straightness or perfection of the flux lines. Also, the rocking-curve-integrated intensity I_{hk} of an (h, k) diffraction spot depends on the square of
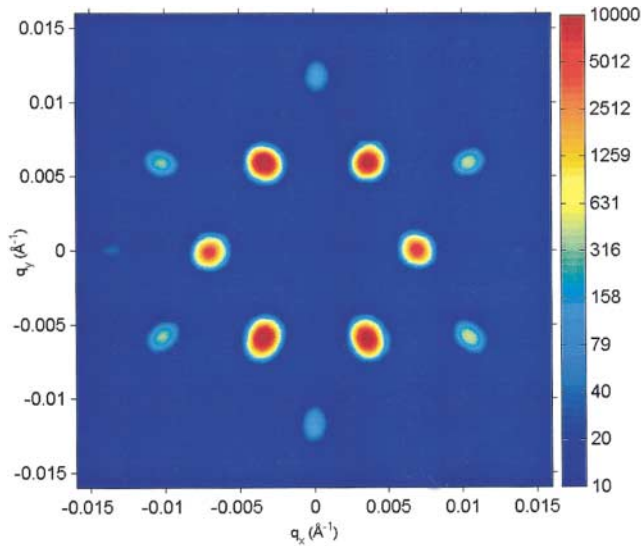
FIG. 1 (color). Diffraction pattern on the SANS multidetector ($128 \times 128 \times 7.5$ mm^2 pixels) from the FLL after cooling in 0.2 T to 3.2 K (logarithmic scale). The picture is a sum of counts obtained with the sample angle rocked either vertically or horizontally to maximize each of the spots in turn. Background scattering, from the sample in the normal state, has been subtracted. A beamstop intercepts the nondiffracted beam.

the Fourier component F_{hk} of the spatial variation of the magnetic field via [12]:

$$I_{hk} = 2\pi V \phi \left(\frac{\gamma}{4} \right)^2 \frac{\lambda_n^2}{\Phi_0^2 q_{hk}} |F_{hk}|^2. \qquad (1)$$

Here, ϕ is the incident neutron flux, γ is the neutron magnetic moment in nuclear magnetons, V is the illuminated sample volume, Φ_0 is the flux quantum, λ_n is the neutron wavelength, and q_{hk} is the magnitude of the scattering vector.

After background subtraction, the position and shape of the $(1, 0)$ spot on the multidetector was fitted by a two-dimensional Gaussian, and the rocking curve was also fitted by a Gaussian. Results of these fits are shown in Figs. 2 and 3. It is clear from the temperature dependence of the larger width (approximately in the tangential direction), that at no temperature (at least on the spacing of our temperature points ∼20 mK) do the spots spread out into a ring as H_{c2} is approached. The slight increase in tangential width at high temperatures corresponds to an angular spread of only 0.9° in FLL orientation. In other respects the FLL becomes *more* perfect near H_{c2}: there is a steady *decrease* with temperature in the rocking curve width and also a small decrease in the radial width of the spot. We have used the method of Ref. [13] and the known incoming beam divergence of 0.0226° and wavelength spread of 4.3% to analyze these changes. The change in radial spot width is a consequence of a change in mosaic spread, which falls from 0.033° at 3.2 K to 0.029° at 5.2 K. We believe that this is due to the nonellipsoidal shape of our sample and incomplete surface pinning of flux lines. This
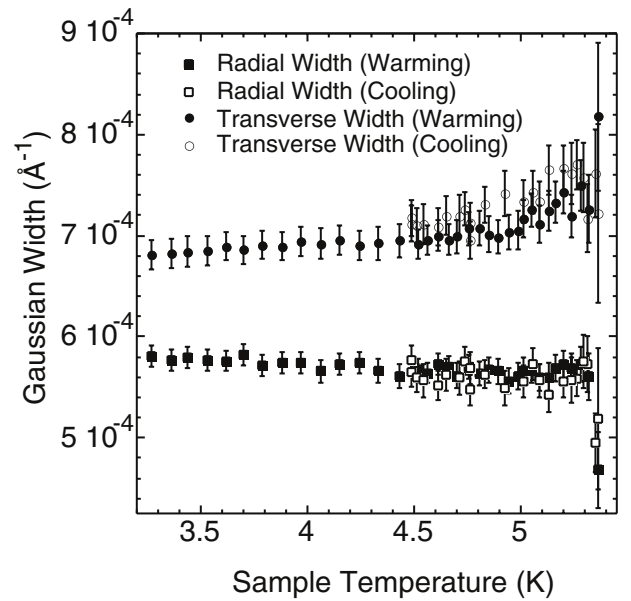

FIG. 2. The major and minor widths of a Gaussian fit to the $(1, 0)$ diffracted spot, obtained at the peak of the rocking curve, versus temperature. Everywhere in this Letter, the quoted "width" of a distribution is the standard deviation, i.e., the rms width.

leads to a slight macroscopic curvature of the lines, which is larger at low temperatures when the sample magnetization is larger. Even at the highest temperature, the mosaic spread of the FLL is nonzero. This may arise from a finite correlation length for the FLL parallel to the field.
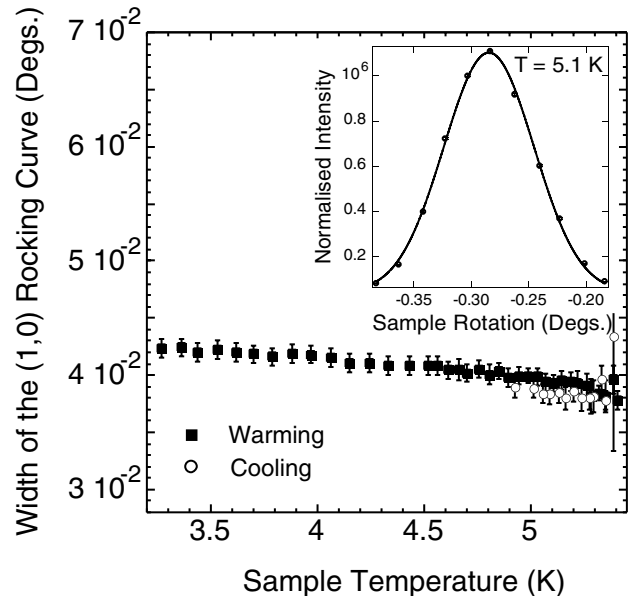

FIG. 3. The width of a Gaussian fit to the rocking curve of the $(1, 0)$ diffracted spot as a function of temperature. The absolute accuracy of the temperature measurement is ∼0.1 K, but the resolution is 10 mK. In the inset is shown a typical rocking curve, taken at 5.1 K, with intensity as a function of the angle of sample rotation about a vertical axis.

We estimate the minimum value, if due to this cause, as $\sim 200 \ \mu$m.

We note also that there is no hysteresis in Figs. 2 and 3. We also checked that a slight oscillation in the value of the external field, which would allow vortices to move into their equilibrium configuration, had no effect. All of these results are in complete accord with the *absence* of a first-order transition or flux lattice melting and in disagreement with the results of Ref. [10]. Further information comes from the temperature variation of the diffracted intensity; as shown in Fig. 4, this goes smoothly to zero when the $H_{c2}(T)$ line is reached at ~ 5.4 K. In Fig. 5, we replot our data near H_{c2} as a square root, so that the ordinate is proportional to $|F_{10}|$ as given by Eq. (1). According to the Abrikosov solution to the Ginsburg-Landau (GL) equations, for example as reported by Brandt [14], both the magnetization M and F_{hk} vary linearly near H_{c2}:

$$M = \frac{-(H_{c2} - H)}{1.16(2\kappa^2 - 1)}; \qquad F_{hk} = (-1)^\nu e^{-\pi\nu/\sqrt{3}} \mu_0 M \,. \tag{2}$$

In this expression, $\nu = (h^2 + hk + k^2)$, and κ is the GL parameter. Using $B = \mu_0(H + M)$, we may eliminate H from Eq. (2) to give

$$F_{hk} = \frac{(-1)^\nu e^{-\pi\nu/\sqrt{3}}(B - B_{c2})}{1 + 1.16(2\kappa^2 - 1)}$$
$$\sim \frac{(-1)^\nu e^{-\pi\nu/\sqrt{3}}(T_{c2} - T)}{1 + 1.16(2\kappa^2 - 1)} \frac{dB_{c2}}{dT} \,. \tag{3}$$

The latter form is appropriate in our case of varying temperature at constant B. It is satisfying to see in Fig. 5 a linear variation of F_{hk} with temperature, as expected from Eq. (3). We also note that within our resolution the line goes linearly to zero, and does not jump to $\sim$ zero intensity, as would be expected at a FLL melting transition.

To make a *numerical* check of these relationships somewhat below T_c, GL theory needs some well-known corrections. There is a temperature dependence to κ [15,16]; also, the magnetization in Eqs. (2) and (3) involves a value κ_2 different from κ_1 that determines the value of H_{c2}. According to magnetization and heat capacity measurements [16,17], κ_2 for the field along the [111] direction for a sample of our purity has the value 1.57 at 5.2 K, compared with 0.73 at T_c. Finally, as calculated by Brandt [18], the ratio between F_{hk} and the magnetization is larger at lower temperatures than that given by Eq. (2). The correction factor at 5.2 K is 1.60. Using $dB_{c2}/dT = -5.2$ mT $\cdot$ K^{-1} at 5.2 K for the field in the [111] direction, the expected value of F_{10} is calculated as $F_{10} = 0.50$ mT. Using Eq. (1), our intensity data at 5.2 K gives the magnitude of F_{10} as 0.50(0.02) mT. To our knowledge, these measurements represent the first *numerical* confirmation by SANS of the field variation in the Abrikosov structure near H_{c2} where the
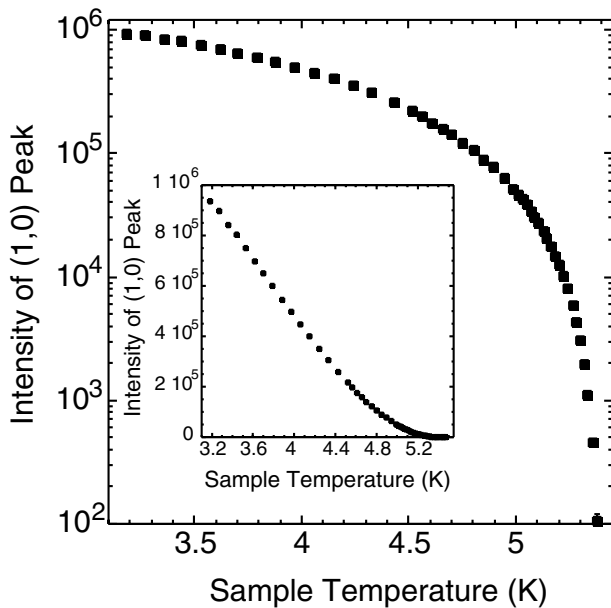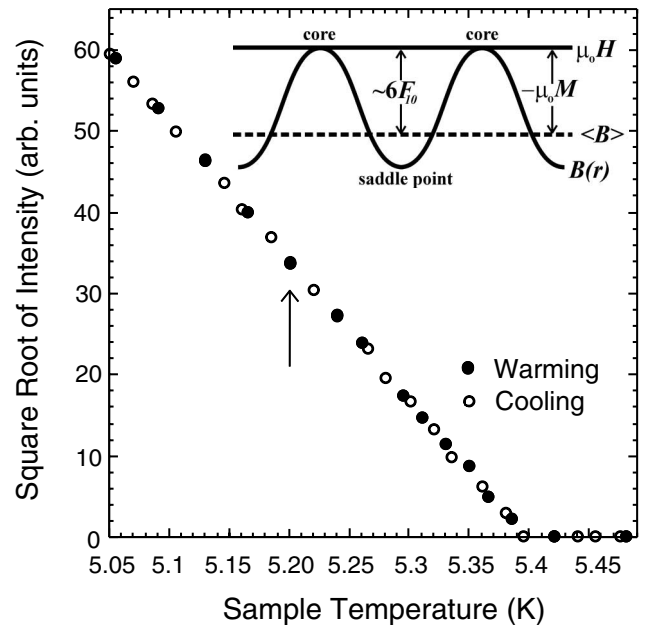

FIG. 4. Background-corrected intensity of the $(1,0)$ spot versus temperature on heating, after cooling in 0.2 T to 3.2 K. The vertical axis is logarithmic to emphasize the dynamic range, $>10^4$, with the lowest counts having an error $\sim 15\%$. In the inset the data are shown on a linear scale.

FIG. 5. The square root of the background-corrected, rocking-curve-integrated intensity of the $(1,0)$ diffraction spot versus temperature in the vicinity of the $H_{c2}(T)$ line. The linear behavior is that expected from Eq. (3). The open points were obtained on heating and the solid points on cooling, showing no hysteresis. At the point marked with an arrow, the field modulation in the mixed state has been calculated from the absolute intensity, as described in the text. The inset represents the field as a function of position along a line joining two flux line cores, showing the interpretation of F_{10}.

diffracted intensity is very small. (Other much earlier data [19] did not give numerical agreement with theory.)

It seems likely that the qualitatively different results reported in Ref. [10] are due to impurity/pinning effects. The values of H_{c2} reported for that sample are almost double those of pure Nb [16], so we can deduce that ℓ is comparable to ξ, increasing κ above the intrinsic value. ac susceptibility studies of that sample show the peak effect, while ours does not under the conditions of our experiment. We conclude that the FLL "melting" below H_{c2} reported in Ref. [10] is most likely a disordering of the flux lines by enhancement of flux line pinning in the peak effect region. Similar disordering transitions have been observed by transport measurements in NbSe$_2$ (doped with Ta to increase the peak effect) [20] and by SANS measurements in (Ba/K)BiO$_3$ [21] and in BiSCCO [4]. The absence of FLL melting well below H_{c2} in Nb is also supported by calculations of the melting line. An equation in a form suitable for Nb is [22]

$$\frac{B_{c2}(T) - B_m(T)}{B_{c2}(0)} = \left(\frac{1.78t}{2\pi c_L^2}\right)^{2/3}(1 - t^2)^{2/3}\mathrm{Gi}^{1/2}, \quad (4)$$

where c_L is the Lindemann ratio, $t = T/T_c$ and $\mathrm{Gi} = \frac{1}{2}[(\mu_0 k T_c)/4\pi B_c^2(0)\xi^3(0)]^2$ is the Ginzburg number, a measure of the strength of thermal fluctuations. Using the known material properties of Nb [17,23], we estimate $\mathrm{Gi} \sim 1.0 \times 10^{-10}$. Using Eq. (4) with $c_L = 0.25$, we estimate the melting line to be ~ 7 mK below the H_{c2} line under the conditions of our experiment. This is consistent with the observation of critical fluctuations in the heat capacity [11] over a similar temperature range near H_{c2}, and our nonobservation by SANS of critical fluctuations or melting at our temperature resolution.

The question remains: How does the hysteretic 1st order transition observed in Ref. [10] evolve from the intrinsic behavior as pinning strength is increased? It is possible that a supposed intrinsic 1st order melting transition in pure Nb is renormalized downwards in temperature, as increased pinning smoothly takes over from thermal fluctuations to disrupt the soft FLL near H_{c2}. In this view, the transition observed in Ref. [10] is the *same* as the melting transition, but into a state so heavily pinned that it appears to have a critical current in ac susceptibility measurements. Alternatively, the transition observed in Ref. [10] is from a topologically ordered FLL into an entangled or dislocated glassy flux solid [6,24] with zero resistivity at small currents. At a higher temperature, this would undergo a vortex glass transition [7] into a liquid state with nonzero resistivity. This question does not appear to be resolved theo-

retically [6,22,25] or experimentally in high T_c materials [26,27]. In niobium, the question may be experimentally resolved by studies including transport measurements as a function of pinning strength.

In conclusion, we suggest that the description "flux lattice melting" should be used to describe situations where the FLL is disrupted predominantly by *thermal* fluctuations. If this condition is adhered to, the "melted" phase will behave as a fluid and will not support a critical current. The "melting" reported in Ref. [10] did not satisfy these conditions. From our observations, we find that in pure Nb the flux lattice structure is stable against thermal fluctuations over essentially all of the mixed state region, and our SANS measurements triumphantly confirm the Abrikosov picture near H_{c2}.

[1] A. A. Abrikosov, Sov. Phys. JETP **5**, 1174 (1957).
[2] D. R. Nelson, Phys. Rev. Lett. **60**, 1973 (1988).
[3] A. Houghton *et al.,* Phys. Rev. B **40**, 6763 (1989).
[4] R. Cubitt *et al.,* Nature (London) **365**, 407 (1993).
[5] A. Schilling *et al.,* Phys. Rev. Lett. **78**, 4833 (1997).
[6] T. Giamarchi and P. Le Doussal, Phys. Rev. B **55**, 6577 (1997).
[7] G. Blatter *et al.,* Rev. Mod. Phys. **66**, 1125 (1994).
[8] T. Natterman and S. Scheidl, Adv. Phys. **49**, 607 (2000).
[9] A. B. Pippard, Philos. Mag. **19**, 217 (1969).
[10] X. S. Ling *et al.,* Phys. Rev. Lett. **86**, 712 (2001).
[11] S. P. Farrant and C. E. Gough, Phys. Rev. Lett. **34**, 943 (1975).
[12] D. K. Christen *et al.,* Phys. Rev. B **15**, 4506 (1977).
[13] R. Cubitt *et al.,* Physica (Amsterdam) **180B&181B**, 377 (1992).
[14] E. H. Brandt, Rep. Prog. Phys. **58**, 1465 (1995).
[15] A. L. Fetter and P. C. Hohenberg, in *Superconductivity* (Marcel Dekker, New York, 1969), Chap. 14; B. Serin, *ibid.,* Chap. 15.
[16] H. R. Kerchner *et al.,* Phys. Rev. B **21**, 86 (1980).
[17] C. G. B. Baker *et al.,* Physica (Amsterdam) **108B**, 927 (1981).
[18] E. H. Brandt, Phys. Lett. A **43**, 539 (1973).
[19] See references in J. Rammer, Physica (Amsterdam) **181C**, 99 (1991).
[20] Y. Paltiel *et al.,* Phys. Rev. Lett. **85**, 3712 (2000).
[21] I. Joumard *et al.,* Phys. Rev. Lett. **82**, 4930 (1999).
[22] G. P. Mikitik and E. H. Brandt, Phys. Rev. B **64**, 184 514 (2001).
[23] D. K. Finnemore *et al.,* Phys. Rev. **149**, 231 (1966).
[24] J. Kierfeld and V. Vinokur, Phys. Rev. B **61**, R14 928 (2000).
[25] T. Reichhardt *et al.,* Phys. Rev. Lett. **84**, 1994 (2000).
[26] N. Avraham *et al.,* Nature (London) **411**, 451 (2001).
[27] K. Deligiannis *et al.,* Phys. Rev. Lett. **79**, 2121 (1997).