ANISOTROPY IN THE FLUX-LINE LATTICE SYMMETRY OF A LOW-κ TYPE-II SUPERCONDUCTOR*

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ABSTRACT

Correlation between the symmetry of the two-dimensional flux-line lattice (FLL) and the real crystal lattice (CL) has been studied in a superconducting niobium sphere by means of small-angle neutron diffraction. A double-perfect silicon-crystal diffractometer enabled precise determination of the three inter-fluxoid distances corresponding to the FFL basic cell.

A systematic study of the anisotropic behavior was made as a function of temperature and magnetic-field amplitude for fields parallel to a few high-symmetry CL axes in the (110) plane. In addition, at T = 4.30 K progressive deformation of the FLL was studied as the sample was rotated in the (110) and (100) planes. The FLL was found to be hexagonal only for fields parallel to the three-fold CL axis. Two-fold symmetry prevailed for other CL directions in these planes except near the four-fold axis, where either of two distorted triangular lattices existed, preserving the reflection symmetry in composite, but not individually. This behavior will be discussed in terms of current models for fluxoid-CL interactions; in particular, in terms of the extension of the Takanaka theory done by Delrieu, Roger, and Kahn.

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INTRODUCTION

The existence of a monocrystal of fluxoids in pure low-$\kappa$ superconductors has been correlated to the anisotropy of the superconducting properties.\(^{(1)}\) The orientation of the basic cell of the flux-line lattice (FLL) with respect to the crystalline lattice (CL) and the shape of this basic cell already have been studied in niobium as a function of the orientation of the applied field, of the field intensity, and of the temperature by Kahn et al.\(^{(2,3)}\), Schelten et al.\(^{(4)}\) and Essman et al.\(^{(5)}\). These previous studies were done with rectangular-shaped samples, and the results may have been influenced by the FLL-surface interaction and by the inhomogeneities of the internal flux density due to non-uniform demagnetization effects.

For these reasons, we used a perfect sphere of pure niobium where both surface and volume pinning have been removed.\(^{(6)}\) A double-perfect single-crystal small-angle diffractometer\(^{(7)}\) enabled the precise determination of the three interfluxoid spacings corresponding to the FLL cell. The sample holder has provision to rotate \textit{in situ} about a well-defined crystalline axis. The precision of 0.3 arc seconds in the measurement of a Bragg diffraction angle in the range of 300 leads to an accuracy of $0.1^\circ$ in the determinations of the angles of the triangular half-cell of the FLL. This accuracy is, in fact, good enough to see the small distortions...
due to the misalignment of the rotational axis with the chosen crystallographic direction. Within the validity of the extension of the Takanaka theory\(^{(8)}\) done by Delrieu, Roger, and Kahn,\(^{(3)}\) a misorientation of 2° of the applied field with respect to the \(<1\overline{1}1>\) axis would yield these typical figures for the three angles of the triangle: 59.9°, 59.6°, and 60.5°. In fact, deviations of this magnitude are observed.

In this paper, we will report the variation of orientation and shape of the FLL basic half-cell:

i) when the applied field sweeps \((1\overline{1}0)\) and \((100)\) planes at constant temperature 4.3 K, at the flux density \(B_0\) of the intermediate mixed state (IMS);

ii) for higher field intensity in the mixed state (MS), especially when the field is parallel to a \(<100>\) and \(<114>\);

iii) when \(T\) is varied between 1.5 and 7.5 K for the \(<1\overline{1}1>\), \(<1\overline{1}0>\), and \(<100>\) directions parallel to the field; this latter orientation has been studied intensively.

**APPLIED-FIELD ORIENTATION DEPENDENCE**

In the Takanaka calculations,\(^{(8)}\) the basic half-cell is assumed always to be an isosceles triangle. Delrieu, Roger, and Kahn\(^{(3,9)}\) pointed out that this is true only when the field is parallel to a symmetry plane of the CL. Throughout this paper, we will compare our experimental results with the predictions of this more general theory, which will be denoted DRK.\(^{(3)}\) For certain CL orientations, the experimental results we obtained exhibit distorted FLL triangular half-cells, even when the field is parallel to such symmetry planes. The symmetry of the physical situation
must be preserved by assuming the possible existence of a second FLL lattice related to the first one according to the reflection symmetry. These two lattices do share the crystal in two domains of equal size. As a matter of fact, the positioning of the sample with respect to the symmetry plane is not accurate enough (± 2°), and this small deviation favors one of these domains. These domains have been observed simultaneously only when the field is nearly parallel to a four-fold axis.

Figure 1 shows what happens when the field scans the (110) CL plane from a <001> to a <110> direction. For H parallel to a <001> axis, there exist two triangular lattices OAB and OA'B'. These half-cells are isosceles with BO = BA and B'O = B'A'. For directions intermediate between the <001> and at approximately the <113>, the half-cell OAB is a distorted (scalene) triangle. At about the <113> orientation, the point B reaches the symmetry <110> axis. At this orientation, the half-cell is nearly isosceles with AO = AB. The isosceles half-cell goes through an equilateral shape for the field parallel to a <111>, and resolves to the isosceles half-cell OA"B" for H parallel to a <110>. The small misalignments and distortions in the last part of the scan (e.g., point "B" should lie on the <110>) are due to the previously mentioned small errors in the field alignment with the designated axis.

Schelten et al. (4) also found two lattices at this orientation, but with bases parallel to the <110>. The shape of these triangles is in agreement with our results.

Figure 2 shows the deformation of the FLL when the field scans the (100) plane from <011> to <001> directions. The basic cell of the FLL glides progressively from the isosceles half-cell OAB (BO = BA) to another
isosceles half-cell OA'B' (A'O = A'B'), instead of the final position OA'B" (very close to the initial OAB), which is also an existing FLL domain when the field is parallel to the <001> axis.

The axis \( \hat{u} \) is the intersection of the plane perpendicular to the field with the (100) cubic face (rotation plane) of the Nb lattice. It marks the symmetry plane, which remains constant all along this process.

In fact, it is not possible to reach the lattice OA'B" by continuous deformation because this FLL domain is not needed to preserve the reflection symmetry, but only the four-fold symmetry specific of the <001> field orientation.

DEPENDENCE ON THE FIELD INTENSITY

It is well established that the hexagonal FLL for \( H \) parallel to a <111> does not change shape or orientation as a function of field intensity. For other directions of field, the change is generally small. For instance, when the field is parallel to <112> Kahn\(^{(3)}\) observed a small variation from 67.6° to 66.8° for the summit angle of the FLL half-cell when the field intensity was varied from 0.5 \( H_{c2} \) to 0.9 \( H_{c3} \). The DRK value is 67.1° in this case.

When the field is parallel to a <100> at 4.3 K, we observed a similar shift, the orientation remaining the same. The summit angle varied from 53.2° at \( H = H_{c1} \) to 54.0° at \( H = 0.7 H_{c2} \) (DRK's value 52.0°). However, at low temperatures, we have strong evidence that a field increase can result in a transformation from the square to the distorted triangular FLL.
Now, when the field is parallel to a \(<114>\), a field increase results not only in a shape change, but also in an orientation shift. As seen in Fig. 3, the scalene triangle which exists at low field has become isosceles at about \(H \sim 0.6 H_{c2}\), and the basis of this triangle is aligned along a \(<110>\) axis; i.e., the FLL then fulfills the reflection symmetry of the CL without need of a second FLL domain. In this case, the summit angle is 69.8°, very close to the DRK prediction of 69.9°.

**TEMPERATURE DEPENDENCE OF FLL SYMMETRY**

In the regime of the attractive fluxoid interaction, the temperature dependence of the FLL symmetry has been studied for fields parallel to the three high-symmetry directions \(<111>\), \(<110>\), and \(<100>\).

As expected, for \(H\) parallel to a \(<111>\), the hexagonal symmetry of the FLL prevails throughout the temperature range (1.6 - 7.5 K).

For \(H\) parallel to a \(<110>\), the half-cell is isosceles, and its shape displays the weak temperature dependence shown in Fig. 4. The experimentally measured anisotropy \(\frac{a}{c} = 1.08\) is much smaller than the predictions of DRK; \(\frac{a}{c} = 1.15\) for 4.3 K. This discrepancy has already been noticed by Kahn.\(^3\) Strictly speaking, the theory is valid only near \(H_{c2}\), while the experiments were performed in the intermediate mixed state at low field. However, the observed field dependence for this direction is very weak. Another discrepancy is the damping of this anisotropy when \(T \to 0\), where the effect should theoretically increase. In principle, anisotropic effects should vanish as \(T \to T_c\), resulting in equilateral FLL for all orientations. From Fig. 4, it is seen that more data are needed at high \(T\) to verify this for \(H\) parallel to a \(<110>\).
For the case H parallel to a $<100>$, the situation is more complex. At temperatures above approximately 4 K, one can always observe two domains of isosceles half-cell FLL. The shape of these cells does approach that of the equilateral half-cell as $T \rightarrow T_c$. Below about 4 K, the half-cell domains distort, but in such a way that the area of the basic cell is nearly the same as that of a square lattice with the same nearest-neighbor distance (Fig. 5a). Finally at $T \leq 2$ K, a structural transition can occur, which results in two domains of square FLL (Fig. 5b). With temperature changes, the deformation of the triangular half-cell is such that the nearest-neighbor direction remains parallel to a CL $<100>$. In the other case, a (nearest-neighbor) side of the square cell is $15^\circ$ off this axis. We did not see any progressive change from one FLL orientation to the other (see Fig. 5).

**COMPARISON WITH CURRENT MODELS**

Several authors have made attempts to explain such orientation and deformation of the FLL. Let us quickly discuss three of them.

H. Teichler$^{(10)}$ developed a nonlocal theory taking into account the anisotropy of the attractive electron-electron interaction, as well as that of the electron band structure. This allows predictions of the orientation of the FLL, but not of the cell shape. The main results are shown in Fig. 6. It does not explain the very particular case where H is parallel to a CL $<100>$.

H. Ullmaier et al.$^{(11)}$ developed a calculation based upon the magneto-elastic interaction between fluxoids and CL. This prediction gives some qualitative information concerning the shape of the basic cell.
Extension of this theory for fields near $H_{c1}$ has been made by Roger.\(^{(3)}\)

Agreement is correct only in the case $T > 2 \text{ K}$ when the field is parallel to a $<100>$, just where other theories fail. However, a calculation of the order of magnitude of the interaction energy due to this effect shows it is always 1000 times less than the other possible effects.\(^{(9)}\)

The theory of Tanaka\(^{(8)}\) considers the anisotropy of the Fermi velocity to derive the order-parameter anisotropy, which can be used to deduce the orientation dependence of the free energy. This free energy leads to the FLL orientation and deformation. DRK\(^{(3)}\) emphasized that it is possible to ignore the specific sources of order-parameter anisotropy, and simply relate the FLL anisotropy to the observed $H_{c2}$ anisotropy. DRK considered two limiting cases near $H_{c2}$.

i) The kernel of the self-consistent Bogoliubov equation of the order parameter is short ranged. This is the case in a clean superconductor near $T_c$. (This case is used for the calculations of this paper at $T = 4.3 \text{ K}$.)

ii) The kernel can be represented as the product of an isotropic function and a function of direction only. This is the case in a clean superconductor at very low temperature.

This theory allows quantitative calculations of orientation and shape of the FLL for any field direction and so offers a better possibility to be checked by experiment. As previously mentioned, this theory predicts larger deformations than observed for fields parallel to a $<110>$, but is in good agreement for the other directions, provided the reflection symmetry is experimentally fulfilled by only one fluxoid domain. In particular, it fails for the $<100>$ where both the predicted orientation and
the shape are wrong. We would like to emphasise that, in this case, the appropriate description may involve higher-order terms in the development of the free energy, which are negligible for other orientations.

It is probably worthwhile to make the necessary improvements on the DRK theory for this special direction, in view of the good agreement obtained for other CL orientations.

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Fig. 1. Progressive deformation of the basic half-cell of the FLL at 4.3 K. The flux density is $B_0$ in the range of the attractive interaction between fluxoids. The applied field $H_a$ sweeps a (110) plane from the <001> to the <110> directions. In the four-fold symmetry two domains exist with isosceles half-cell OBA and OB'A' (with half-cell angles 63.5°, 63.3°, 53.2°) where the bases are parallel to <100> axes. During the sweep, the point B follows the path B B'' as the point A glides towards A''. When B is off the symmetry axis, the half-cell is a scalene triangle. After the orientation <113> has been reached, the cell remains isosceles with its basis parallel to <110> axis as predicted by DRK. The path BB'' does not follow exactly the <110> at this stage because of small misorientations of the field with respect to the symmetry plane.

Fig. 2. Progressive deformation of the basic half-cell of the FLL at 4.3 K. The flux density is $B_0$. The applied field sweeps a (100) plane between the <011> and <001> directions. During this process, the direction $\vec{u}$, marking the symmetry plane, is not a constant indexed direction. The field parallel to <011> yields the OAB basic half-cell, which is an isosceles triangle (62.3°, 62.3°, 55.4°) with base parallel to $\vec{u}$. When the field is parallel to the <001>, a possible lattice is the isosceles half-cell 0 A"B" (63.4°, 63.3°, 53.3°), which is very close to OAB. In spite of this possibility and of DRK's prediction, the actual
path goes towards the second possible lattice OA'B'. Every intermediate position is a scalene triangle, and there does exist a second lattice to preserve the symmetry with respect to $\hat{u}$.

Fig. 3. The <114> direction has been found to be unique when the flux density is $B_0$ inside the sample. As the field is increased up to $0.6 H_{C2}$, the basic cell rotates and changes its shape. At this latter value of the field, the cell is isosceles ($55.1^\circ$, $55.0^\circ$, $69.8^\circ$) with its basis along a <110> axis. DRK predicts ($55.03^\circ$, $55.03^\circ$, $69.93^\circ$), in very good agreement with the higher-field results.

Fig. 4. Temperature dependence of the ratio of the leg $a$ to the basis $c$ of the FLL half-cell when field is parallel to <110>. This ratio is expected to be unity at $T = T_c$ (equilateral triangle), 1.15 at $T = 4.2$ K according to DRK, and larger if $T \to 0$. The solid line is used as a guide for the eye.

Fig. 5. Temperature dependence of the shape of the FLL half-cell for fields parallel to a <100> CL axis.

a) The variation of the ratio of height $Y_2$ to basis $X_1$ of the triangle points out the easy transformation from an isosceles to a half square at constant area (i.e., constant flux density) when the temperature is below 4 K.

b) The variation of the ratio of the two legs over the basis points out the departure from the isosceles shape and also the discontinuity in the transformation from the triangle to
the half square. This transition in the shape occurs at the same temperature as the jump in the orientation, with the so-called basis suddenly rotating by 15°.

Solid lines are used as guides for the eye.

Fig. 6. Summary of the publication of the theories of (1) Teichler, (2) DRK, (3) Ullmaier et al. near $H_c^2$, and (4) Ullmaier and Roger near $H_c^1$, compared to the experimental results of this work.
NIOBium

$H_a$ in (110) Plane

$T = 4.300 \, K$

$B = B_0$ (IMS)

Fig. 1
NIOBIUM
$H_0$ IN (100) PLANE
$T = 4.300 \text{ K}$
$B = B_0$ (IMS)
NIOBIUM

Nb - ND1

T = 4.3 K

H = II (114)

\[
\begin{array}{cccc}
\text{H_a} \text{(Oe)} & \psi_1 & \psi_2 & \psi_3 \\
1165 & 68.2^\circ & 57.5^\circ & 54.3^\circ & 10^\circ \\
1646 & 69.8^\circ & 55.15^\circ & 55.05^\circ & 2.2^\circ \\
\end{array}
\]

Fig. 3
NIOBIUM

Fig. 4
Fig. 5
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Fig. 6