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### Fluxoids and neutron polarisation effects

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#### Abstract

There are several ways of investigating flux lines with polarised neutrons. We shall describe our recent work on IN15 at ILL, investigating spatially varying fields perpendicular to the fluxoid axes, which are present with tilted fields in anisotropic superconductors. These field components may be detected by polarisation analysis of neutrons diffracted by the flux line lattice, and allow a detailed investigation of flux structure in such materials as YBCO and NbSe<sub>2</sub>. A further application of polarised neutrons is the use of neutron spin-echo techniques to measure the speed of moving flux lines. We have recently demonstrated this in an Nb–Ta alloy on instruments IN11 and IN15, by measuring the energy change of neutrons diffracted by flux lines, which are moving under the influence of a transport current. We shall also comment on others' work on the search for interference terms between flux lattice and nuclear diffraction signals, and the use of neutron depolarisation for investigation of flux distributions in superconductors. © 1999 Elsevier Science B.V. All rights reserved.

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# 1. Introduction: Polarisation analysis of neutron diffraction by flux lines

In this paper we shall describe four different applications of polarised neutrons to the investigation of

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flux lines, using techniques, invented and developed by others. We shall deal with each separately, beginning with polarisation effects in neutron diffraction by flux lattices in anisotropic superconductors.

The use of small-angle neutron scattering (SANS) to investigate the flux line lattice (FLL) in anisotropic superconductors has, over recent years, led to enormous insight into the vortex behaviour in these systems [e.g. Refs. [1,2]]. For instance, the

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intensity of diffraction can be related to the (temperature-dependent) penetration depth, the shape of the flux lattice reveals the anisotropy of the interactions between fluxoids, the orientation of flux lattice planes may reflect intrinsic processes, or pinning of flux lines to defects such as twin planes, and the thermal vibrations of flux lines may lead to flux lattice melting. On the IN15 instrument at the ILL, we have recently measured polarisation-analysed SANS from the vortex lattice [3] - a technique that is able to reveal yet more information on flux lattice structure. For a conventional isotropic superconductor in the mixed state, the local flux density inside the bulk of the sample always lies parallel to the applied magnetic field. Hence, neutron scattering occurs without flipping of the spins of the neutrons [4]. However, in an anisotropic superconductor - for example in the layered High- $T_{\rm c}$  materials, this may not be the case, since the supercurrents will tend to flow in the CuO<sub>2</sub> planes, which will not be perpendicular to the average induction if the field is applied at an angle to the c-direction. This gives rise to spatially varying local fields which have components perpendicular to the direction of the average induction (which, by flux quantisation, is also the direction of the flux line axes). Transverse components of the local field will flip the neutron spins from parallel to antiparallel to the field, or vice versa [4], and are therefore detectable by polarisation-analysed SANS. We believe that until our experiment, these transverse components of local magnetic field had not been previously detected by any technique. For instance, they are not observable in macroscopic measurements since they average to zero over the unit cell, so there is no net magnetisation.

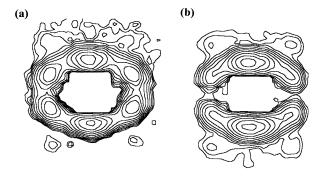


Fig. 1. Scattered intensity from the FLL formed in untwinned YBCO with an induction of 0.5 T applied at  $55^{\circ}$  to c: (a) intensity of non-spin-flipped neutrons, (b) spin-flipped.

If the value of magnetic field inside a high- $\kappa$ superconductor is not close to  $B_{c2}$  (which condition is easily satisfied in high- $T_c$  materials!), the interaction between the cores of the flux lines may be neglected, and the London model may be applied for the variation of magnetic field near a vortex. An important property of layered superconductors is the anisotropy of the magnetic field penetration depth. We may describe the magnitude of this anisotropy by a quantity  $\gamma$ , which represents the ratio of the penetration depth due to supercurrents flowing in the c-direction to that for currents flowing in the basal (CuO<sub>2</sub>) planes. In YBCO,  $\gamma$  typically has a value 5–8, although in other high- $T_c$  materials, it may be larger than 100. In the anisotropic London model, the superconducting electrons are described by an effective mass tensor, and the effective mass ratio is equal to  $\gamma^2$ . In the simplest approximation, we may take the basal plane to be isotropic (and at sufficiently low fields, we may ignore any d-wave

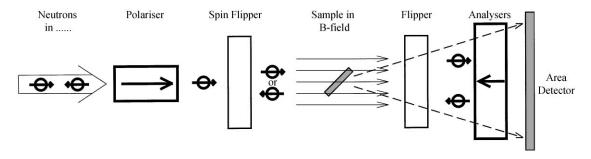


Fig. 2. Schematic arrangement of IN15 used to measure the data shown in Fig. 1.

effects on FLL structure), so with the field applied perpendicular to the planes, the FLL should have a regular hexagonal unit cell. When the average induction is at an angle  $\theta$  to the c-axis, the flux lattice then should be a distorted hexagon, and the distortion may be represented by a "stretching" of both real lattice and diffraction spots (which are an image of the reciprocal lattice) [5]. If the induction is not too close to  $B_{c1}$ , the six diffraction spots lie on an ellipse of axial ratio given by

$$(\min \text{or/major}) = (\cos^2 \theta + \gamma^{-2} \sin^2 \theta)^{1/2}. \tag{1}$$

It will be seen that for large  $\gamma$ , and  $\theta$  not too close to 90°, this expression reduces to  $|\cos\theta|$ , so that for instance with  $\alpha=45^\circ$ , the hexagonal lattice is distorted by a factor  $\sim 1/\sqrt{2}$ . This distortion (combined with a small amount of basal plane anisotropy) may be seen in the diffraction patterns in Fig. 1. The anisotropy also gives a preferred orientation to the flux lattice: the orientation seen in Fig. 1, with a pair of diffraction spots parallel to the rotation axis, is that predicted from theory [5].

Thiemann et al. [6] have calculated the Fourier components, in three dimensions, of the local flux density in an anisotropic vortex lattice described by the London model with a uniaxial effective mass tensor. They define a set of Cartesian axes in the following way: z is along the direction of the average induction B (perpendicular to the page in Fig. 1.); y is the axis about which B is rotated from c (vertical in Fig. 1) and x lies in the plane of rotation (horizontal in Fig. 1). In general, there will be spatially-varying local field components  $b_x$ ,  $\boldsymbol{b}_{v}, \boldsymbol{b}_{z}$ :  $\boldsymbol{b}_{z}$  will cause diffraction without spin-flip and the other two components (which are present for all  $\theta$  except  $0^{\circ}$  and  $90^{\circ}$ ) will cause spin-flip. Now for magnetically ordered systems, the intensity of diffraction has a "moment orientation factor", which leads to zero intensity when the moments are parallel to the scattering vector. This arises because the magnetic field due to the moments has zero divergence. In the present case,  $\operatorname{div} \boldsymbol{b} =$ 0 leads to the following condition for the amplitude of b at a particular FLL reciprocal lattice vector g:

$$\mathbf{g} \cdot \mathbf{b}(\mathbf{g}) = 0. \tag{2}$$

Since all reciprocal lattice vectors g are perpendicular to z, the non-flip diffraction by  $b_z$  is unaffected by condition (2). However, when g lies in the x-direction,  $b_y$  is zero by symmetry, and  $b_x$  is zero from (2). Hence, no spin-flip scattering is expected for diffraction spots lying in the x-direction (horizontal in Fig. 1); however, for any g having a nonzero  $g_y$ , diffraction with spin-flip is expected.

The experimental arrangement of IN15 for these experiments is shown in Fig. 2. Technical details of IN15 will be found in Ref. [7]. Longitudinally polarised neutrons pass through a spin flipper, are diffracted by flux lines created in a sample with a longitudinal magnetic field, and pass through another flipper, followed by analysers in front of a multidetector. A longitudinal guide field is applied to maintain neutron polarisation between these components. The analysers absorb neutrons with the initial polarisation, so in the ideal case, with both flippers off ("-" configuration), only spin-flipped neutrons would reach the detector, and with either flipper on (+ - or - +) non-spinflipped neutrons would pass. The finite efficiency of polariser, analyser and flippers was measured, at each pixel of the detector, by placing a pyrolytic graphite (non-spin-flip) scatterer behind the sample in the normal state, and recording the neutron counts for all four flipper configurations in turn (--, +-, -+ and ++). Measurements on the sample were made in the same configurations, both in the presence of the flux lattice, and also in the normal state, to allow subtraction of the background scattering from sample and cryostat. Using the measured apparatus efficiencies, results from the - and + configurations were analysed to give the non-spin-flip and spin-flip diffraction from the flux lattice. In Fig. 1 are shown typical results, taken with the field at 55° to the c-axis. The intensities of different diffraction spots were in excellent qualitative agreement with the London theory predictions [6], and showed the expected "field-orientation" effect. However, there systematic differences between were results, and those obtained from the -+ and + + configurations. This effect of the initial neutron polarisation may indicate that the sample was behaving in a chiral manner, or may be due to a yet

undiscovered systematic error. In the near future, we intend to check this behaviour by also investigating the more conventional layered superconductor NbSe<sub>2</sub>, which will provide a control. In addition, we intend to make further measurements on YBCO at higher fields. This is of great interest, because when the field is raised, the orientation of the flux lattice alters by 90°, to that not expected from London theory [5]. The polarisation-analysis technique is showing great promise, and should allow us to check the flux-line/flux-line interactions with unprecedented sensitivity.

## 2. Interference between flux lattice and nuclear scattering

If there is, in addition to the effect of magnetic field on neutron spin, a non-magnetic amplitude to the scattering from a flux lattice – for instance if the flux line cores have a different density from the rest of the material, then interference between the magnetic and non-magnetic terms will lead to a dependence of the scattered intensity on spin direction. This process is analogous to the well-established technique of measuring induced magnetisation densities, by measuring the flipping ratio of Bragg peaks, which arise from a combination of nuclear and magnetic scattering. This effect was recently reported to occur in the FLL in Nb [8].

However, there is serious doubt, on numerical grounds, that the observed effect can be due to a change in density in the cores of the flux lines. The ratio between FLL intensities with the neutron spin parallel and antiparallel to the field direction was observed to differ from unity by a fraction  $\sim 1.5 \times 10^{-2}$ . This was interpreted as a ratio between nuclear and magnetic scattering lengths  $\sim 4 \times 10^{-3}$ , which may be modelled as a fractional change in density of the flux line cores,  $\varepsilon \sim 3 \times 10^{-4}$ . If this occurs, there will be a consequent storage of elastic energy U per unit volume in the cores, given by

$$U = \frac{1}{2}C\varepsilon^2,\tag{3}$$

where the relevant modulus C for Nb  $\sim 2.5 \times 10^{11}$  Pa, which gives the elastically stored energy  $\sim 10^4$  J m<sup>-3</sup>. Is this large or small? To decide, we compare it with the condensation energy due to

superconductivity, which is equal to  $1/2\mu_0H_c(T)^2$ . At 4.2 K,  $\mu_0 H_c \sim 0.16$  T [9], giving a condensation energy also  $\sim 10^4$  J m<sup>-3</sup>. This figure clearly rules out strains comparable with those suggested in Ref. [8]: any deformation of the vortex cores must be a small effect consequent on the formation of the superconducting state, and make a small contribution to the total energy. If this were not the case, the pressure-dependence of  $T_c$  would be much larger than observed. It is clear that whatever the cause of the results reported in Ref. [8], they are not due to lattice deformation. Incidentally, it should be pointed out that X-ray diffraction is a better technique to detect lattice deformations, since it is directly sensitive to density changes, and has a far higher flux. Such an X-ray experiment has been attempted on a high- $T_c$  superconductor [10], and has not shown effects comparable with those claimed for Nb.

## 3. Measurement of flux line motion by spin echo techniques

There is a great deal of interest in the effects of flux line motion on flux structures and melting [e.g. Refs. [11,12]]. Flux line motion, induced by a large enough transport current, averages and reduces the effect of pinning and is expected to create a moving Bragg Glass [12,13]. At the time of writing, there has been no confirmation of these ideas by SANS in high- $T_{\rm c}$  materials; there is some indirect evidence from decoration measurements in NbSe<sub>2</sub> [14].

Many years ago, it was demonstrated that it is possible to measure flux line motion by SANS [15]. In these experiments, the change in Bragg angle for diffraction from the moving lattice was observed. It is also possible to measure the change  $\Delta \varepsilon$  in energy of the neutrons on diffraction: this may easily be calculated as the energy change due to reflection from a moving mirror:

$$\Delta \varepsilon = h v_{\rm ff} / d,\tag{4}$$

where  $v_{\rm ff}$  is the speed of the flux lattice planes of spacing d. The speed may be related to the induction B and driving electric field E via:

$$v_{\rm ff} = E/B. \tag{5}$$

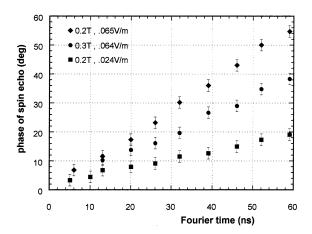


Fig. 3. Spin-echo phase shift versus Fourier time for various values of E/B in an Nb–Ta sample. Experiments performed on IN11 and IN15 at ILL.

For typical values of  $v_{\rm ff}$  of  $< 1~{\rm m~s}^{-1}$ ,  $\Delta \varepsilon$  is a very small energy and can only be detected by the neutron spin-echo technique. Recent experiments on IN11 and IN15 in a low- $T_{\rm c}$  sample [16] have demonstrated the effect. An energy change  $\Delta \varepsilon$  in the scattered neutrons causes a phase shift  $\phi$  in the spin-echo which is given by:

$$\phi = \frac{\Delta \varepsilon}{\hbar} t = \frac{2\pi v_{\rm ff}}{d} t,\tag{6}$$

where t is the Fourier time of the precession, which is proportional to the line-integral of the spin-echo precession field. Typical results are given in Fig. 3, and show that the average speed of the flux lines may be measured by this technique; this quantity is more easily and cheaply measured with a voltmeter using Eq. (5)! However, with this technique it is also possible to measure the distribution of vortex speeds in a specimen [16].

# 4. Investigation of flux structures with neutron depolarisation

Another way to investigate magnetic field distributions is to investigate the change in polarisation of neutrons transmitted through the sample. If the field in the sample is uniform, then all transmitted neutrons travelling at a given speed will have their degree of polarisation unchanged (although the direction of polarisation may have altered, due to the Larmor precession of the neutron spins during the time they are in the field). On the other hand, if the field is non-uniform, then the beam may be depolarised. This technique, relying on transmission, rather than scattering, is clearly sensitive to field variations over distances rather larger than the flux lattice spacing, and can therefore be used to investigate flux gradients, pinning etc. In the general case, the incident neutron polarisation can be in any direction relative to the incident neutron beam, so the technique is referred to as "three-dimensional neutron depolarisation" (3DND), and has application to magnetic materials ([17] and M.Th. Rekveldt, this workshop proceedings) as well as superconductors [18].

The theory may be simplified as follows [19]. We suppose that there is a small region in the sample, of dimension  $\zeta$ , in which the field deviates from the average field and has an extra component  $B_n$  perpendicular to the neutron spin direction. This region can cause depolarisation if the apparent turning frequency of the field ( =  $v/\zeta$ , where v is the speed of the neutron) is larger than the Larmor frequency,  $\gamma B$ . This condition is usually satisfied inside a sample (e.g. up to  $\zeta = 0.2$  mm for  $\lambda = 1.5$  Å neutrons at B = 100 mT), so the neutron spin cannot adiabatically follow the field direction and in the time  $(\zeta/v)$  precesses by a small angle  $\phi =$  $\gamma B_{\rm n}(\zeta/v)$  about the local field. As a result the neutron polarisation is reduced by the factor  $\cos \phi \sim 1 - \phi^2/2$ . If there are n such independent regions along the neutron path, then the depolarisations add in a random walk fashion and the net polarisation becomes

$$P = \exp(-n\gamma^2 \langle B_n^2 \rangle \zeta^2 / 2v^2)$$
  
=  $\exp(-\gamma^2 \langle B_n^2 \rangle \zeta \cdot t / 2v^2),$  (7)

where  $t = n\zeta$  is the thickness of the sample.

One possible ND setup would be with the initial neutron spin and average magnetic field parallel to the neutron beam: in that case, if bundles of flux lines of dimension  $\zeta$  are tilted with respect to the average field direction, then depolarisation will

occur [19]. Alternatively, with longitudinally polarised neutrons, the field may be aligned perpendicular to the neutron beam. In this case, the average field causes an overall rotation of the polarisation but in addition, either tilts of flux bundles or variations of the magnitude of the field will cause depolarisation. It should be mentioned that variations of magnetic field around an individual flux line occur on the length scale of the flux line spacing, d. These variations will give rise to SANS if the flux lines are oriented correctly for diffraction. They will not give rise to detectable depolarisation unless the detector has sufficient aperture to accept the diffracted beams, and even in this case the effects are often small [20]. This may be regarded as arising because the length scale  $\zeta \sim d$  is too short. Thus SANS and neutron depolarisation may be regarded as complementary: one is sensitive to flux lattice structure and the other to longer-range variations of magnetic field.

Three-dimensional neutron depolarisation measurements may be used in two different ways. Firstly, one may use the fact that the transmitted beam has its overall polarisation rotated: this may be detected by 3d analysis of the transmitted neutrons, which is carried out in any case to measure the depolarisation. With the incident neutron spin aligned perpendicular to the field in the sample region, the spin rotation gives a measure of the integral of the magnetic field along the neutron path. If the value of this integral is too large, so that the neutron velocity spread causes too much depolarisation, the average field in an unmagnetised sample may be compensated by an equal and opposite field in the neutron path outside the sample region. In this case, any additional rotation is proportional to the magnetisation of the sample if the neutron beam and field are exactly aligned [17]. In general, one also has to worry about the effects of stray fields outside the sample. However, if the sample magnetisation is perpendicular to the beam, the stray fields may be removed from the beam by connecting a magnetic voke to the ends of the sample [see e.g. Ref. [20]]. The rotation technique may be used in tomographic mode by restricting the neutron beam to a small aperture: a positional resolution of 0.2 mm is possible [17,21], without excessive loss of intensity. By this means, demagnetising effects of flat plates and remanent trapped fields have been measured as a function of position in High  $T_{\rm c}$  samples.

Alternatively, depolarisation measurements can give information about variations in field magnitude inside a sample. However, Eq. (7) tells us that we must model the field distribution inside the sample in order to obtain detailed information, because the depolarisation depends on the product of field variation  $\langle B_n^2 \rangle$  and correlation length  $\zeta$ . In particulate magnetic materials, microscopy may be able to provide the value of  $\zeta$ , so that definite conclusions may be drawn [17]. In superconductors, one must assume the dimensions of "flux bundles" or the value of  $\langle B_n^2 \rangle$  in order to make further progress. The most definite results that have been obtained so far are in Refs. [18–21].

In principle, 3DND gives a large amount of information: a 3 × 3 "depolarisation matrix" relating each component of the input polarisation to the output polarisation. Also, since it uses the straight through beam, rather than scattered neutrons, it is not so limited in intensity as many polarised neutron techniques. Depolarisation is sensitive to tilts of flux lines through much larger angles than SANS would be able to measure, and can be used to detect variations in magnetic field over larger distances than SANS (either via the dependence of the results on  $\zeta$ , or by the scanning tomographic technique). However, the need to model the results has meant that very little completely new information has been obtained on high-T<sub>c</sub> materials - and occasionally [22] the interpretation has been extremely speculative and probably wrong! 3DND is a technique that still has to prove its utility in superconductivity research.

#### 5. Summary

We see that polarised neutrons can contribute in at least two ways to the investigation of flux lines in superconductors. There is no doubt that polarisation analysis of diffracted neutrons can give information about magnetic field distributions in anisotropic superconductors. Furthermore, spinecho techniques can be used to measure flux line motion in appropriate cases. However, if there is a density change of the superconductor in the cores of flux lines, then small-angle X-ray diffraction is the preferred technique, since measurements of spin-orientation-dependence of diffracted intensity cannot possibly obtain the same sensitivity. Finally, neutron depolarisation techniques are certainly sensitive to flux line distributions, but the analysis of the results is model dependent, so may not give as clear a result as non-neutron techniques.

**Note added in proof:** Following comments from F. Tasset and C. Fermon at this meeting, and further experiments, it has become clear that the unexpected features of the polarisation analysis results were mostly due to subtle effects of the Zeeman splitting of the neutron energy in the *B*-field of the flux lattice: (P.G. Kealey et al., to be published).

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