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Title:	Direct	t physical observation of theFFLO state in CeCoIn5					
<b>Research area:</b>	Physic	S					
This proposal is a	resubn	nission of 5-42-544					
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Samples: CeCo	oIn5						
Instrument			Requested days	Allocated days	From	То	
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Abstract:							

A famous prediction (by Fulde & Ferrell and Larkin & Ovchinnikov: FFLO) is for a spatially-modulated superconducting state in any material where the effect of high magnetic fields on the antiparallel spins of electrons in Cooper pairs places an upper limit on the field at which superconductivity can occur (¿Pauli-limited superconductivity¿). There have been various indirect, macroscopic indications of the FFLO state in Pauli-limited materials but no direct observation, and suggestions that an analogous state can occur in cold atom condensates. The FFLO state should appear below the upper critical field at low temperatures. In the more stable LO version, it contains planes of zeroes of the superconducting order parameter. These planes are perpendicular to the magnetic field, and slice through the flux line lattice (FLL) which is also present. The nonzero momentum of the Cooper pairs represented by this spatially-modulated state arises because the two halves of a Cooper pair have different Zeeman energy in the magnetic field, and therefore must have a compensating difference in their kinetic energy and momentum. We propose to search for FFLO in CeCoIn5 using a novel SANS geometry.

## Experimental Report: Direct physical observation of the FFLO state in CeCoIn<sub>5</sub>

**Abstract:** An additional spatial variation of superconductivity (the FFLO state) is predicted to occur in Paulilimited superconductors near the upper critical field at low temperatures. We used theoretical predictions of the variation of the FFLO wave-vector and SANS intensity versus field to search for this phenomenon, which is very difficult to observe directly, although there are many indirect experimental data suggesting the presence of this state when the field direction is close to or along the *c*-axis [1-4]. After a very careful search, we found no evidence for the FFLO state in CeCoIn<sub>5</sub> with the field close to the *c*-axis. We conclude that either this state does not exist in CeCoIn<sub>5</sub>, or that the predictions of intensity or wavevector direction and magnitude are not correct, or that pinning gives a wide range of FFLO *q*-values, which would give too wide a rocking curve to observe the FFLO intensity.

**Experimental geometry:** We set up our sample so that in the field range of interest it would have flux line lattice (FLL) diffraction spots in the horizontal direction. The FFLO satellites – arising from planes of zero energy gap  $\Delta$  - are predicted to be separated along the field direction from the FLL spots - as shown in Fig. 1. Clearly, a



horizontal-field magnet is required, with wide windows allowing the incoming and outgoing neutron beams to be at a large angle to the field. This was achieved using 258OXHV49, which has  $\pm -22 \deg$ windows. However, they are necessarily of aluminium, which gives larger background than quartz or silicon windows.

When the field is along the crystal c-direction, there are two slightly-distorted hexagonal FLL domains tied to the tetragonal crystal axes. We rotated our sample - a mosaic of single crystals - by 15 deg about the vertical axis so that the whole sample was occupied by a single FLL domain which had spots in the horizontal direction, as observed in Fig. 2. (The magnet was rotated to bring just one spot to the Bragg condition.)



15° to c (only the RHS spot on the Bragg condition)

The predictions for the intensities of the FLL spot and the FFLO satellites are shown in Fig. 4 on the following page. We first established the variation of FLL intensity with field, as shown in Fig. 3. The intensity does not vary as sharply with field as predicted in Fig. 4(b), but





we expect that the FFLO state will occur in the region where the intensity is falling, and we concentrated on this field range.

We see from Fig. 4(a) that the FFLO q is expected to vary rapidly with field, rising from zero at the onset field. The satellites will occur at 2q, as there are two zeroes of  $\Delta$  per wavelength. As illustrated in Fig. 1, a satellite at 2q would satisfy the Bragg condition at a different rock angle than that for the FLL. Therefore, so we rocked over a wide range of angles to search for the signal over a wide range of q. Fig. 4(c) shows the predicted FFLO intensity at 2q, which should be largest just above the onset field and about  $1/30^{\text{th}}$  of the FLL signal there. As indicated in Fig. 3, we were not certain about the value of the onset field, so we carried out a two-dimensional search - in field and rock angle looking for the weak FFLO signal.

Fig. 1 shows that if there is an FFLO signal on one side of the detector at a particular rock angle, there should be another on the other side of the detector at a somewhat different rock angle. The difference will be  $2\theta_{\text{satt}}$ , where  $\theta_{\text{satt}}$  is the Bragg angle for the satellites, which is approximately the same as the FLL Bragg angle. This allowed us to discriminate between apparent intensity due to Poisson statistics and real signals. In our search for FFLO signals, we tested different collimation settings to establish that we were maximising signal to background noise. We also we changed to a second mosaic, which had an orientation rotated by 45 deg around the *c*-axis. This allowed us to bring the other FLL spots close to the horizontal axis. These spots might behave differently, as they are not aligned with the directions of the nodes in the *d*-wave pairing of CeCoIn<sub>5</sub>. In all these cases, we saw no significant FFLO signal. A typical rocking scan in

shown in Fig. 5. Many of these could have been shown! We conclude that either this state does not exist in CeCoIn<sub>5</sub>, or that the predictions of intensity or wavevector direction and magnitude are not correct, or that pinning

gives a wide range of FFLO *q*-values, which would give too wide a rocking curve to observe the FFLO intensity.

## References

[1] S.-Z. Lin *et al.*, Phys. Rev. Lett. **124**, 217001 (2020).

[2] K. Kumagai *et al.*, Phys. Rev. Lett. **97**, 227002 (2006).

[**3**] A. Bianchi *et al.*, Phys. Rev. Lett. **91**, 187004 (2003).

[4] J. S. White *et al.*, New J. Phys. **12**, 023026 (2010).
[5] K. M. Suzuki *et al.*, J. Phys. Soc. Jpn. **80**, 123706 (2011).



Fig 5: Typical FFLO satellite search. Box sums at L & R predicted FFLO positions *vs* magnet rotation angle