Theory of Neutron Diffraction from the Vortex Lattice in UPt$_3$

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Abstract

Neutron scattering experiments have recently been performed in the superconducting state of UPt$_3$ to determine the structure of the vortex lattice. The data show anomalous field dependence of the aspect ratio of the unit cell in the B phase. There is apparently also a change in the effective coherence length on the transition from the B to the C phases. Such observations are not consistent with conventional superconductivity. A theory of these results is constructed based on a picture of two-component superconductivity for UPt$_3$. In this way, these unusual observations can be understood. There is a possible discrepancy between theory and experiment in the detailed field dependence of the aspect ratio.

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Experiments of the last several years on superconducting UPt₃ have revealed a rich phase diagram. The phase boundaries have been mapped with considerable accuracy by locating anomalies in specific heat [1], ultrasound [2], and torsional oscillator signals [3]. The additional transitions found may be explained by assuming that the superconductivity is unconventional, and that the order parameter has two complex components [4]. At zero pressure, there are three superconducting phases, called the A phase (low fields and high temperatures), the B phase (low field, low temperature), and the C phase (high field, low temperature). The theoretical explanation remains untested, however, unless detailed experimental characterization of the phases can be carried out, preferably by searching for definite signatures of the predicted phases. The theory suggests that the B phase is the most promising in this regard. In this phase, the two components are predicted to coexist, and their interplay can give rise to novel effects.

The comparison of theory and experiment is not straightforward, however, as no microscopic experimental probe couples directly to the order parameter. Because of this, it is desirable to have a probe for which predictions can be made using the phenomenological Ginzburg-Landau theory alone, thus circumventing microscopic details. Neutron diffraction from the flux lattice depends only on the field distribution in the sample [5] and therefore may be calculated from the phenomenological theory, yet it offers a level of detail which cannot be obtained by any other means [6]. A new experiment of this kind has recently been carried out and is reported in the accompanying Letter [7].

Calculations of the lattice structure and the field distribution have so far been limited to the neighborhood of the upper critical field $H_{c2}$ [8] [9] [10]. However, it is only at low temperatures that the penetration depth is short enough that experiments can be done. In practice this means well away from $H_{c2}$. Furthermore, the interesting effects occur in the field dependence of the scattering intensities in the B phase, and particularly near $H^*$, the transition field from the B to the C phase. The purpose of this Letter is to present calculations of the lattice structure and the scattering cross section as a function of applied field, and to compare the results to experiment.
The scattering amplitude for a change in neutron wavevector $\vec{Q}$ in the Born approximation is

$$f_B(\vec{Q}) = \frac{im_N\mu_N}{\hbar^2(2\pi|\vec{k}_i|)^{1/2}} \int d^2r h(\vec{r})e^{i\vec{Q}\cdot\vec{r}} = \frac{im_N\mu_NA}{\hbar^2(2\pi|\vec{k}_i|)^{1/2}} h(\vec{Q}),$$

(1)

where $m_N$ and $\mu_N$ are the mass and magnetic moment of the neutron and $\vec{k}_i$ is the incoming wavevector. $h(\vec{r})$ is the field in the sample and $h(\vec{Q})$ is its Fourier transform. The integration is over a cross-section $A$ of the sample perpendicular to the beam. The object of interest, the observed intensity $I(\vec{Q})$, is thus proportional to $|h(\vec{Q})|^2$. The experiments have all been performed with $\vec{H}$, the applied field, directed in the basal plane, so the calculations presented here will be only for that field direction. I deal only with Bragg scattering, so $\vec{Q}$ is a reciprocal lattice vector.

If the $y$-axis (in the basal plane of the hexagonal crystal) is taken as the field direction, two-component theories have the generic free energy density:

$$f(\vec{\eta}) = \alpha(|\eta_x|^2 + |\eta_y|^2) + \beta_1(\vec{\eta}\cdot\vec{\eta}^*)^2 + \beta_2|\vec{\eta}\cdot\vec{\eta}|^2$$

$$+ K_x|D_x\eta_x|^2 + K_y|D_x\eta_y|^2 + K_z(|D_z\eta_x|^2 + |D_z\eta_y|^2).$$

(2)

(3)

$\eta_x$ and $\eta_y$ are the two components of the order parameter. $D_x \equiv -i\partial_x - 2eA_x/\hbar c$ and similarly for $D_z$. $\eta_x$ and $\eta_y$ may belong to one of the two-dimensional representations $E_1$ or $E_2$ of the point group, or they may belong to different, but accidentally nearly degenerate representations. The scattering experiments probably do not distinguish these possibilities. For simplicity, I have taken $\eta_x$ and $\eta_y$ to be degenerate (only a single $\alpha$) at zero field. This is reasonable at the low temperature (50 mK) at which the experiments are done. In fact I shall fix the temperature and treat $\alpha$ as constant. This means that only deal with the B (low field or $H < H^*$) and C (high field or $H > H^*$) phases, and the transition between them, will be discussed. The free energy $F = \int f[\vec{\eta}(\vec{r})]d^3r$ where $f[\vec{\eta}(\vec{r})]$ is given by Eq. 2 does indeed lead to the observed phase diagram with three superconducting phases for $\vec{H}$ in the basal plane. The form of $\vec{\eta}(\vec{r})$ in the C phase is known. It is found that $\vec{\eta}(\vec{r}) = (\eta_x(\vec{r}), 0)$. Thus the free energy reduces to
The problem represented by this free energy density is isomorphic to that of an s-wave superconductor with mass anisotropy. One may therefore transcribe well-known results [12]:

\[
|h(Q)|^2 = H^2 \left[ 1 + (\lambda_z C)^2 Q_z^2 + (\lambda_x C)^2 Q_x^2 \right]^{-2} \exp \left\{ -\frac{1}{2} \left[ (\xi_x C)^2 Q_x^2 + (\xi_z C)^2 Q_z^2 \right] \right\},
\]

where \(\xi_i C = (K_i/\alpha)^{1/2}\) are the coherence lengths and \(\lambda_i C = (\hbar^2 c^2/32\pi e^2 K_i |\eta|)^{1/2}\) are the penetration depths in the C phase. Since the free energy density may be transformed to the isotropic form by rescaling coordinates: \(x' = (\xi_z/\xi_x)^{1/2} x\) and \(z' = (\xi_x/\xi_z)^{1/2} z\), the fluxons form a rescaled hexagonal lattice (centered rectangular lattice). In London theory the vortex cores act as delta-function sources of the field. Correct ions to this require numerical calculations which have been carried out by Brandt [13], and it is concluded that the sources are well represented by Gaussians - this is reason for the exponential factor in Eq. 5. The coordinate scaling determines the opening angle \(\alpha_L\) of the reciprocal lattice (defined in Fig. 1), which uniquely determines the aspect ratio of the unit cell. It is given by \(\tan^2(\alpha_L) = 3K_x/K_z\). This result and flux quantization allow us to calculate the reciprocal lattice vectors as a function of field. Substituting these values into Eq. 4 shows that \(I(Q)\) is the same for all \(Q\) in the first shell (the six smallest nonzero \(Q\)). These are the only points measured to date.

Theory thus predicts that three properties qualitatively characterize the C phase:

(i) the lattice structure (shape of the unit cell) is independent of field;

(ii) all peaks in the first shell have the same intensity;

(iii) the intensities fall off exponentially with field, with \(-d \ln I/dH \sim \xi_x C \xi_z C\).

Property (i) has been pointed out before [14], [10]. The C phase has no unique signatures of unconventional superconductivity, however, since an s-wave superconductor with mass anisotropy has all of these features.

The B phase is quite different. I first develop the theory and then turn to comparison with experiment.
The opening angle in the B phase can be computed in the regime where \( \xi_i \ll a \ll \lambda_i \), where \( a \) is the lattice constant for the vortices. In terms of the field, this is \( H_{c1} \ll H \ll H_{c2} \). Since UPt\(_3\) is strongly type-II, this is a substantial range. It will suffice for our purposes to compute the currents at a distance of order \( a \) from the cores, because the momentum transfers of experimental interest are of order \( 1/a \). Thus the structure of the cores at short distances of order \( \xi_i \), a complex subject, is not of interest here. At these larger distances the \( \beta_2 \) term in Eq. 3 locks the relative phase of \( \eta_x \) and \( \eta_y \): \( \eta_y = \pm ir(H)\eta_x \), where \( r(H) \) is a real ratio. \( r(H) \) supplies the interesting field dependence in the B phase. Since \( \tilde{\eta}_y \) appears continuously, \( r(H) \) is a nonnegative, monotonically decreasing function of \( H \) with \( r(H^*) = 0 \). The phase-locking relation, together with the free energy of Eq. 3, gives a London equation for the currents with the penetration depths

\[
(\lambda_x^B)^2 = \frac{\hbar^2 c^2}{32\pi e^2 |\eta_x|^2 [K_x + r^2(H)K_y]},
\]

and

\[
(\lambda_z^B)^2 = \frac{\hbar^2 c^2}{32\pi e^2 |\eta_x|^2 K_z [1 + r^2(H)]}.
\]

In the field regime under consideration, these currents determine the lattice structure and one finds

\[
\tan^2(\alpha_L) = \frac{K_x + r^2(H)K_y}{3[1 + r^2(H)]K_z}.
\]

The shape of the unit cell is strongly field-dependent in the B phase. This effect does not occur in one-component superconductors, conventional or unconventional.

The computation of the intensities in the B phase is not so straightforward, since corrections to London theory are involved. However, the same phase-locking approximation allows us to make a mean-field-type theory. We neglect amplitude correlations in the region where the distance from the cores is much greater than the coherence lengths. Then the effective field on either component has the same spatial dependence as in the s-wave case, and we may again apply the results of Brandt. The complication which arises is that both
\( \eta_x \) and \( \eta_y \) act as sources of the field. This leads to separate exponential dependences, and the breakdown of the simple relationship \(-d \ln I/dH \sim \xi^2\). There are now two effects which determine the field dependence of the intensity. First, the interaction of \( \eta_x \) and \( \eta_y \) given by the quartic terms in Eq. 3 sets up effective fields which give a field dependent renormalization of the correlation lengths of both components. This means that the exponents acquire an additional field dependence relative to Eq. 5. Second, the prefactors of the exponents have a field dependence because of the separate contributions from \( \eta_x \) and \( \eta_y \), whose relative weight is field-dependent.

The full expression for the \( h(\vec{Q}) \) may be separated into a part which depends relatively weakly on \( H \) and is probably unobservable, and the exponential part \[1\]. The full expression, and details of its derivation, will be given elsewhere. The exponential part is

\[
h(\vec{Q}) \sim [(1 + r^2)^{-1} F\left(\frac{\xi_B}{\xi_{xx}}, \xi_{xx} Q_x, \xi_{zz} Q_z\right)]
\]

\[
+ (1 + r^2 \frac{K_y/K_x}{n})^{-1} F\left(\frac{\xi_B}{\xi_{xx}}, \xi_{xx} Q_x, \xi_{zz} Q_z\right) \times \exp\left[\left(-(\xi_{xx}^2 Q_x^2 - (\xi_{zz}^2 Q_z^2)/4\right)\right] \quad (10)
\]

\[
+ [(1 + r^{-2})^{-1} F\left(\frac{\xi_B}{\xi_{yy}}, \xi_{yy} Q_x, \xi_{zy} Q_z\right)]
\]

\[
+ (r^{-2} + K_x/K_y)^{-1} F\left(\frac{\xi_B}{\xi_{yy}}, \xi_{yy} Q_x, \xi_{zy} Q_z\right) \times \exp\left[\left(-(\xi_{yy}^2 Q_x^2 - (\xi_{zy}^2 Q_z^2)/4\right)\right] \quad (12)
\]

In this formula, the coherence length \( \xi^{B}_{xx} \) is given by

\[
\xi^{B}_{xx} = \left(\frac{K_x}{\alpha}\right)^{1/2} \left\{1 + \frac{1}{4}\left[(1 - \frac{\beta_2}{\beta_1})(2 - \frac{H}{H_{c2}} - \frac{H}{H_y}) - \frac{1}{4}\left[(1 - \frac{\beta_1}{\beta_2})(\frac{H}{H_{c2}} - \frac{H}{H_y})\right]\right]\right\}^{1/2}, \quad (13)
\]

and there are similar formulas for the other three coherence lengths. The field \( H_y \) is a constant given by

\[
H_y = \frac{H^*}{1 - b + bH^*/H_{c2}}, \quad (14)
\]

with \( b \equiv (\beta_1 - \beta_2)/(\beta_1 + \beta_2) \), while the function \( F(x, q_1, q_2) \) is defined as

\[
F(x, q_1, q_2) = x \int_{-\infty}^{\infty} du \int_{-\infty}^{\infty} dv \frac{u^2}{x^2 u^2 + v^2} \exp\left[-(u - i q_1/2)^2 - (v - i q_2/2)^2\right]. \quad (15)
\]
Thus the effective coherence length, if it is defined by the slope of \( \ln I(\vec{Q}) \) as a function of field, is seen to be field dependent. This again is utterly characteristic of a multicomponent system, and cannot be found in a one-component superconductor.

These calculations have been done assuming that the separation of the singularities in \( \vec{\eta}_x \) and \( \vec{\eta}_y \) in the unit cell are separated by a distance much less than the lattice constant. If this is not the case, as in the 'shifted' phase predicted near the tetracritical point, then the intensities for the reciprocal lattice vectors in the first shell may be quite different because of extinction effects. In particular, at low field, the intensity for \( \vec{Q}_{0,1} \) will be less than that for \( \vec{Q}_{1,-1} \). Unfortunately, explicit calculations for this phase are quite difficult.

Eqs. 8 and 12 give three qualitative predictions for the B phase:
(i) the lattice structure (shape of the unit cell) is depends on field;
(ii) peaks in the first shell have different intensities;
(iii) the intensities have a complex field dependence, with deviations from the exponential shape;
(iv) there is a kink in \( I(H) \) at the second-order B-C transition. Property (i) has been pointed out before [9], [14], [10]. Property (iv) does not imply that the BC transition is first-order. The slope of \( I(H) \) is not interpreted as proportional to a single coherence length, (which would then be discontinuous). Instead, the kink is interpreted as signalling the continuous growth of a new component of the scattering. The BC transition is second-order in this theory.

Before comparing these predictions quantitatively with experiment, we must discuss the determination of the Ginzburg-Landau parameters. The final results for the opening angle of the flux lattice depend only on the ratio \( \beta_1 / \beta_2 \) and the ratios of the stiffness constants \( K_x, K_y \) and \( K_z \). I take \( \beta_1 / \beta_2 = 0.5 \), as determined by specific heat experiments. The stiffness constant ratios as determined by fitting to the neutron data are \( K_x : K_y : K_z = 1.5 : 0.88 : 2.5 \).

The opening angle is plotted in Fig. 1. The theory predicts a field-dependent lattice
structure in the B phase: \( H < H^* = 5.3kOe \), as is observed. There should be a kink in the curve at \( H^* \) and in the high-field C phase, \( \alpha_L \) should be independent of field. These predictions are consistent with the data, but more points at higher fields and smaller error bars are needed to provide a real test.

The logarithm of the form factor \( H_1 \) at the Bragg point \( \vec{Q}_{1,1} \) is plotted as a function of field in Fig. 2 with the same parameter ratios \( K_x : K_y : K_z = 1.5 : 0.88 : 2.5 \). The coherence length, defined as the geometric mean of the coherence lengths in the \( x \) and \( z \) directions, was taken to be 101 Å. Very good agreement with experiment is obtained. In particular, the truly novel feature in the data, the kink at \( H = H^* = 5.3kOe \), is very well reproduced. I have also calculated the intensity at the Bragg point \( \vec{Q}_{0,1} \). It is not shown because for the parameter range here the calculated intensities differ by only a few per cent. It will be difficult to verify that this difference exists at the current level of experimental accuracy.

The stiffness constant ratios obtained by the present fit can be compared to ratios obtained by fitting to the measured values of the upper critical fields at temperatures near \( T_c \) for different field directions and from the discontinuity in slope at the tetracritical point. This gives \( K_x : K_y = 2 \), and applying constraints from particle-hole symmetry yields \( K_x : K_y : K_z = 1 : 2 : 4 \). It is quite clear that the two methods disagree, and even the ordering differs between \( K_x \) and \( K_y \). Indeed, if \( K_x < K_y \), as suggested by the critical field slopes, the curve \( \alpha_L(H) \) is monotonically decreasing for \( H < H^* \), in clear contradiction to the data of Fig. (1). The significance of the discrepancy is not clear at present, since it involves an extrapolation from high to low temperatures which may not be justifiable. Nonlocal corrections at low temperatures are surely important and the nature of these corrections for unconventional superconductors is not known.

The qualitatively new features predicted for neutron scattering from the flux lattice in the B phase are observed, as is the relatively conventional behavior of the C phase. Both the opening-angle data and the field-dependent intensities can be fit to good accuracy. This fit does not agree with one obtained from critical field measurements. This may be due to difficulties of extrapolation to the low temperatures of the experiment, or may indicate a
real discrepancy between theory and experiment.

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FIGURES

FIG. 1. Theoretical curve and experimental data points from Ref. 8 for the opening angle of the flux lattice as a function of the applied field. The inset shows the definition of the angle.

FIG. 2. Theoretical curve and experimental data points from Ref. 8 for the logarithm of the form factor at the first Bragg peak as a function of the applied field.
$\ln (H_1)$ (arbitrary units) vs. $H$ (kOe)