Inhomogeneous Magnetic Phases: A Fulde-Ferrell-Larkin-Ovchinnikov-Like Phase in Sr$_3$Ru$_2$O$_7$

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The phase diagram of Sr$_3$Ru$_2$O$_7$ contains a metamagnetic transition that bifurcates to enclose an anomalous phase with intriguing properties—a large resistivity with anisotropy that breaks the crystal-lattice symmetry. We propose that this is a magnetic analogue of the spatially inhomogeneous superconducting Fulde-Ferrell-Larkin-Ovchinnikov state. We show—that through a Ginzburg-Landau expansion where the magnetization transverse to the applied field can become spatially inhomogeneous—that a Stoner model with electronic band dispersion can reproduce this phase diagram and transport behavior.

Fulde and Ferrell [1] and Larkin and Ovchinnikov [2] conjectured that the transition between a superconductor and insulator, driven by a magnetic field, could occur via an intermediate phase with spatially modulated superconducting order. This proposal has since been extended to a wide range of settings, from ultracold atomic Fermi gases [3] and exciton insulators [4] to quark matter and neutron stars [5]. However, experimental confirmation of these predictions is still controversial [6,7]. In a similar spirit, intermediate phases between a Fermi liquid and Wigner crystal [8] have been discussed. We propose an inhomogeneous magnetic phase that can be considered a magnetic analogue of the LOFF phase. In this case, a change in homogeneous ferromagnetic order occurs via an intermediate phase with spatially modulated magnetization. This phase would generate clear experimental signatures. Furthermore, we argue on the basis of both new and previous experimental results that the anomalous phase behavior observed in Sr$_3$Ru$_2$O$_7$ [9–13] can be explained in this way.

The bilayered ruthenate Sr$_3$Ru$_2$O$_7$ shows a sequence of metamagnetic transitions [12]. Recent angle-resolved photoemission spectroscopy (ARPES) data have found evidence of van Hove singularities that may drive this metamagnetism [14]. Early studies focused on a line of metamagnetic critical end points that could be tuned to a quantum critical point by adjusting the magnetic field strength and orientation [13]. Subsequently, ultrapure samples showed a bifurcation of this line upon approaching the putative quantum critical point [9,10] with a second line of critical end points emerging from the zero-temperature plane (see Fig. 1). This bifurcation is accompanied by the appearance of a striking peak in resistivity [9] with curious, anisotropic dependence on the relative orientation of current, lattice, and in-plane magnetic field [11]. When current flows in the crystallographic direction most parallel to the in-plane field, the resistivity peak rapidly decreases as the field is moved away from the c axis. When it is nearly perpendicular to the in-plane field, the peak persists. Further indications of a “roof” delineating the region of anomalous phase behavior with field along the c axis [9] were provided by a kink in the longitudinal magnetization and a qualitative change in the temperature dependence of resistivity. Figure 1 uses new resistivity data to extend this roof in angle. Similar features occur elsewhere in the phase diagram [11], with further bifurcations apparent upon approaching the ab plane. These show a smaller resistance anomaly, but have the same characteristic anisotropy.

Beginning with a heuristic discussion of the physics of the LOFF state and its magnetic analogue, in the following, we will describe how the Wohlfarth-Rhodes [15] band picture of metamagnetism is extended to allow the possi-
bility of spatially modulated magnetic phases. In order to deduce the effects upon the broader phase diagram, we turn to a Ginzburg-Landau expansion of the microscopic Hamiltonian. The key physics is revealed in an expansion along the line of metamagnetic critical end points through a vanishing stiffness to spatial modulation of the transverse magnetization. This leads to a reconstruction of the phase diagram. Finally, we describe how our picture explains the behavior of Sr$_3$Ru$_2$O$_7$—capturing both the experimental phase diagram and the properties of the anomalous phase.

A BCS superconductor is formed by binding electrons at the Fermi surface with opposite spin and momentum ($\mathbf{k}, \uparrow$ and $-\mathbf{k}, \downarrow$) to form Cooper pairs. A magnetic field imposes a Zeeman energy cost on the superconductor which is balanced against the condensation energy. When Zeeman energy dominates, the superconducting state is destroyed; Cooper pairs break, allowing a spin polarization to develop. The transition from superfluid to normal phase can occur via an intermediate inhomogeneous condensate, the LOFF phase [1,2]. By pairing electrons into a state with nonzero total momentum ($\mathbf{k} + \mathbf{q}/2, \uparrow$ and $-\mathbf{k} + \mathbf{q}/2, \downarrow$), the reduction in condensation energy due to modulation is offset by a gain in Zeeman energy. The precise texture of the superconducting order depends sensitively upon microscopic details [5].

A similar mechanism can apply to itinerant magnets: A spatially-modulated magnetic phase may intervene between the high- and low-magnetization states of a metamagnet. To form a ferromagnet, there must be an energetic gain in transferring an electron from a spin-down to a spin-up state of the same momentum. In a Stoner model, this is due to Coulomb exchange energy acquired at the expense of kinetic energy. Extending the Stoner model to include a dispersion with peaks in the electronic density of states (DOS) leads to metamagnetism [15,16]: As the Fermi surface of, say, majority carriers approaches van Hove filling, the single-particle energy cost in changing its filling is reduced. This can lead to a step change in the magnetization at certain values of the external field.

Inhomogeneous magnetic states can be stabilized by DOS peaks in a similar way to spin density waves [17,18]. The simplest such phase formed from a ferromagnet is a spin spiral [19]. A spiral of the right wave vector is reduced. This can lead to a reconstruction of the phase diagram and the properties of the anomalous phase. Setting $\mathbf{M}/\bar{M} = (1 + \phi(\mathbf{r}))\mathbf{e}_\parallel + \phi_\perp(\mathbf{r})$, where $\bar{M}$ denotes the mean-field magnetization along the metamagnetic line [22], and substituting into (1) gives

$$\frac{\beta F_L}{\hbar M} = -H\phi + R\phi^2 + \frac{5}{8}\phi^4 + \frac{1}{2}(1 - \phi + \phi^2)\phi_\perp^2 - \frac{1}{8}\phi_\perp^4 + \cdots$$

(2)

FIG. 2 (color). (a) Energy contours for a next-nearest neighbor tight-binding model with Fermi surfaces for minority and majority electrons shown in blue and red, respectively, (left). The minority and majority bands are distorted by a spiral modulation with nonzero transverse magnetization and wave vector (right). (b) DOS of minority and majority states with a uniform magnetization in magnetization density, $\mathbf{M}$, as [20]
$H$ and $R$ parameterize deviations from the metamagnetic critical end point perpendicular and parallel to the first-order line. The dependence of the higher order coefficients on $H$ and $R$ can be neglected. Although we are interested in reconstructions of the metamagnetic transition that may lie outside the radius of convergence of the parent Landau theory (1), it turns out that an explicit expansion for the Stoner model gives the same coefficients as (2) when constrained to lie along the parent line of critical end points [23].

To allow for inhomogeneous phase formation, we consider a minimal gradient expansion of the free energy:

$$\beta F_{GL} = \beta F_L + (K_\perp + K_1 \phi + K_2 \phi^2 + K_3 \phi^3)(\nabla \phi)^2 + L_\perp (\nabla^2 \phi)^2,$$

where the parameters $K_1$, $K_2$, $K_3$, and $L_\perp$ are functions of the external parameters fixed by the microscopic theory. We neglect terms involving gradients of $\phi$. While such terms can lead to spatial modulation, they do not lead to the phase reconstruction that we find. Gradient terms of fourth order and higher ought strictly to respect the lattice anisotropy [24]. We consider the isotropic case for simplicity.

The key ingredient introduced by explicit evaluation of the gradient expansion for the Stoner theory—that cannot be anticipated on purely symmetry grounds—is that $K_\perp$ changes sign along the line of metamagnetic critical end points [25]. This indicates an instability to the formation of spiral transverse magnetization. As this spiral order is established, the effective $\phi^4$ term changes sign leading to a tricritical point [10].

The resulting phase diagram is shown in Fig. 3. The metamagnetic sheet bifurcates at a dislocated (symmetry broken) tricritical point as shown in green [10]. The bifurcated wings embrace a region of inhomogeneous transverse magnetization in accord with our heuristic description. This region is further enclosed by a surface of continuous phase transitions, shown in blue, at which the transverse magnetization falls to zero. The longitudinal magnetization shows a kink on this surface—a ghost of the transition in the transverse magnetization.

The inhomogeneous magnetic structure may consist of a superposition of several wave vectors. The sum of these must be zero to avoid a spontaneous spin current. A fourfold lattice symmetry (as in Sr$_3$Ru$_2$O$_7$) suggests four preferred wave vectors. There are two ways to superpose these: in pairs of $\pm q$ leading to a spin density wave in one of two directions that breaks the fourfold rotational symmetry to $2$; all four symmetry related wave vectors leading to a spin crystal which preserves the lattice symmetry. An example of the latter is shown in Fig. 3.

Comparison with the experimental phase diagram, Fig. 1, is obtained by expressing $R$, $H$, and $K_\perp$ as functions of the experimental parameters $T$, $\theta$, and $h$. These functions are analytic (as is confirmed by their microscopic calculation) and in the usual spirit of the Ginzburg-Landau expansion their leading dependence near to the critical point is linear [26]. We expand about the point along the line of metamagnetic critical end points where $K_\perp$ changes sign. Figure 4 shows the result of such a correspondence. The natural parameters of our microscopic theory are field, temperature and band filling. An additional mechanism is required to map from filling to angle. One candidate is spin-orbit coupling [27] (which leads to an angle dependent Zeeman coupling) together with orbital effects of an in-plane field. The anomalous behavior of Sr$_3$Ru$_2$O$_7$ is sensitive to disorder as it only appears in the cleanest samples. Our mechanism shows this sensitivity, since disorder smooths out features in the DOS.

Spatially inhomogeneous magnetic structures lead inevitably to enhanced scattering in certain directions. In order to fully explain the anisotropy, there must be a mechanism for an in-plane magnetic field to align the magnetic inhomogeneity. Our simple model does not contain such a mechanism. We suggest that its origin lies in a modification to the dispersion due to in-plane magnetic field, which breaks the symmetry between different orientations of the underlying helices. In the anomalous phase

![FIG. 3 (color). Phase diagram for the Ginzburg-Landau theory with possible spin texture. Green sheets represent first-order transitions in $\phi$. Blue sheets represent continuous transitions into the inhomogeneous phase. The possible spin texture is constructed from four spin helices arranged in a square. The longitudinal magnetization has been suppressed in this picture for emphasis.](image1)

![FIG. 4 (color). The phase diagram rotated into the experimental orientation. The dashed line shows a trajectory through the inhomogeneous region. The inset shows the variation of longitudinal and transverse magnetization through this trajectory.](image2)
this magnetic inhomogeneity leads to enhanced resistivity. With a magnetic field in the \( c \) direction, the inhomogeneity does not break the crystal symmetry (at least macroscopically) and resistivity is isotropic. As the field is rotated into the plane, the magnetic inhomogeneity no longer preserves the lattice symmetry—either through the formation of an anisotropic spin crystal or by a preponderance of domains of spin density waves of one orientation. This anisotropy is reflected in resistivity.

Spatial modulation of magnetization should show up as Bragg peaks in elastic neutron scattering in the anomalous region. Unfortunately, no such data exist. There are, however, pseudelastic data outside of the anomalous region consistent with fluctuations that would freeze into the type of spin crystals that we predict [28].

The mechanism of inhomogeneous magnetic phase formation presented here contrasts with two other proposals: (i) Spin-orbit interactions in systems without a center of inversion symmetry lead to a Dzyaloshinskii-Moriya interaction [29] that favors the formation of magnetic spirals [30] and possibly magnetic crystals [31]. We restrict attention to systems, such as \( \text{Sr}_3\text{Ru}_2\text{O}_7 \), that have a center of inversion symmetry. (ii) Analysis of quantum fluctuation corrections to the theory of itinerant magnets suggests that they can induce metamagnetism and magnetic inhomogeneity [32]. Whether such effects are important in \( \text{Sr}_3\text{Ru}_2\text{O}_7 \) is unclear. We expect that van Hove singularities are characterized by larger energy scales and provide the dominant mechanism. Others have speculated that the anomalous phase in \( \text{Sr}_3\text{Ru}_2\text{O}_7 \) may be a nematic metal with a modulation presented here contrasts with two other proposals:

In conclusion, it has long been established that the Stoner model with a peak in the DOS can lead to metamagnetism. We have shown that a generic band dispersion leads to a bifurcation of this metamagnetism by the intervention of a phase of spatially modulated magnetism analogous to the superconducting LOFF state. This behavior might have been seen already in \( \text{Sr}_3\text{Ru}_2\text{O}_7 \). Indeed, our analysis is rather general and its results may have broader applicability, e.g., \( \text{NbFe}_2 \) [34] exhibits a peak in resistivity associated with the bifurcation of a metamagnetic transition and finite wave vector magnetic order and \( \text{ZrZn}_2 \) [35] may show similar features.

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[19] \( u(k + q/2, j) + u(k - q/2, j) \) with \( u^2 + v^2 = 1, \) transform to real space states with Euler angles \( \theta = \cos^{-1} u, \phi = \mathbf{q} \cdot \mathbf{r} \).


[22] In the Stoner model, \( M = 2h/g + \sum_{\sigma=\uparrow,\downarrow} \sigma n_F(\epsilon_k - gM\sigma/2) \), where \( n_F(\epsilon) \) denotes the Fermi distribution.


[24] In the 2D square lattice, the isotropic contribution to the Ginzburg-Landau free energy density, \( (\tilde{\sigma}^2\phi)^2 \), is augmented by a term proportional to \( (\tilde{\sigma}^2\phi)^4 \).

[25] \( K_1 = -\sum_{k\sigma} (\sigma n_F(\epsilon_{k,\sigma}) + gM\sigma n_F(\epsilon_{k,\sigma}))/2|\partial_k \epsilon_k|^2, \) where \( n_F' = \partial' n_F \) and \( \epsilon_{k,\sigma} = \epsilon_k - gM\sigma/2 \).

[26] While our microscopic theory can be used to determine the dependence upon external parameters beyond linear order, the electron dispersion is not known with sufficient accuracy for this to be useful here. We will discuss these nonlinear dependencies in a forthcoming longer paper.


[31] For a recent review see, e.g., R. Casalbuoni and G. Nardulli, Rev. Mod. Phys. 76, 263 (2004).


