

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

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The phase diagram of $\text{Sr}_3\text{Ru}_2\text{O}_7$ 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—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.

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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 $\text{Sr}_3\text{Ru}_2\text{O}_7$ [9–13] can be explained in this way.

The bilayered ruthenate $\text{Sr}_3\text{Ru}_2\text{O}_7$ 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-

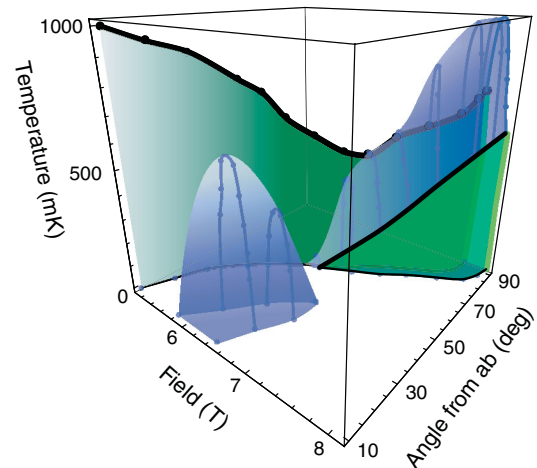


FIG. 1 (color). The phase diagram of $\text{Sr}_3\text{Ru}_2\text{O}_7$ as inferred from in-plane transport properties. The green planes correspond to abrupt changes in resistivity as a function of field. Blue shading indicates regions where the in-plane resistivity is anomalously high, becomes highly anisotropic with respect to the in-plane component of the field [11], and shows an anomalous temperature dependence. The phase diagram obtained from magnetic susceptibility [10] shows the same first-order transitions as indicated here in green, but lacks the roof.

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 $\text{Sr}_3\text{Ru}_2\text{O}_7$ —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 distorts the Fermi surface so that some regions are brought closer to van Hove filling (see Fig. 2). The reduction in single-particle energy cost due to occupying states near to the peak in the DOS can outweigh the single-particle energy costs from elsewhere. This leads to peaks in the transverse magnetic susceptibility [17] and provides a mechanism by which a metamagnetic transition can split, the transition between low and high magnetization occurring *via* a phase of inhomogeneous transverse magnetization.

Such behavior can be shown explicitly for a Stoner model with band dispersion: $\hat{\mathcal{H}} = \sum_{\mathbf{k}, \sigma=\uparrow, \downarrow} \epsilon_{\mathbf{k}} \hat{n}_{\mathbf{k}, \sigma} - U \hat{n}_{\uparrow} \hat{n}_{\downarrow} - \mu_B H (\hat{n}_{\uparrow} - \hat{n}_{\downarrow})$, where $\hat{n}_{\mathbf{k}, \sigma}$ is the number operator for electrons with momentum \mathbf{k} and spin σ and $\hat{n}_{\uparrow(\downarrow)}$ is the

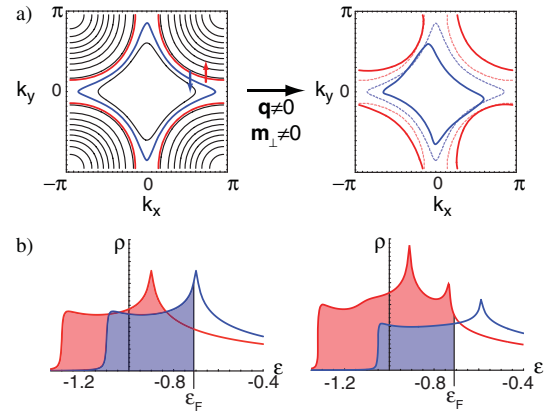


FIG. 2 (color). (a) Energy contours for a next-nearest neighbor tight-binding model with Fermi surfaces for minority and majority electrons shown in blue and red, respectively, (left). The minority and majority bands are distorted by a spiral modulation with nonzero transverse magnetization and wave vector (right). (b) DOS of minority and majority states with a uniform magnetization (left) and with a spiral distortion (right). In the former, the Fermi surface lies just below a peak in the DOS and in the latter it lies between two split peaks.

total number operator for spin-up(down) electrons. $\epsilon_{\mathbf{k}}$ is the electronic dispersion—we focus, without loss of generality, upon a two-dimensional tight-binding dispersion with next-nearest neighbor hopping. As noted above, this model displays metamagnetism [15,16].

Inhomogeneous phase formation leads to a reconstruction of the phase diagram that is best revealed through a Ginzburg-Landau expansion. The thermodynamic properties of a metamagnet can be developed as a Landau expansion in magnetization density, \mathbf{M} , as [20]

$$\beta F_L = r\mathbf{M}^2 + u\mathbf{M}^4 + v\mathbf{M}^6 - \mathbf{h} \cdot \mathbf{M}, \quad (1)$$

where $\mathbf{h} = h\hat{\mathbf{e}}_{\parallel}$ is the external magnetic field. The coefficients of this expansion for the Stoner model may be calculated by a perturbative expansion in interaction [21], and are specific functions of the external parameters: magnetic field, temperature and distance from van Hove filling. The point $r = u = h = 0$ is the parent tricritical point where the line of continuous Stoner transitions at zero field ($r = h = 0, u > 0$) bifurcates symmetrically into two lines of metamagnetic critical end points parameterized by the conditions, $\partial_M F_L = \partial_M^2 F_L = \partial_M^3 F_L \stackrel{!}{=} 0$.

As we are interested in a reconstruction of the metamagnetic transition, we shift our expansion from zero magnetization to an expansion about the magnetization along the line of metamagnetic critical end points. Setting $\mathbf{M}/\bar{M} = (1 + \phi(\mathbf{r}))\hat{\mathbf{e}}_{\parallel} + \phi_{\perp}(\mathbf{r})$, where \bar{M} denotes the mean-field magnetization along the metamagnetic line [22], and substituting into (1) gives

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

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:

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

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 ϕ . 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 ϕ^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

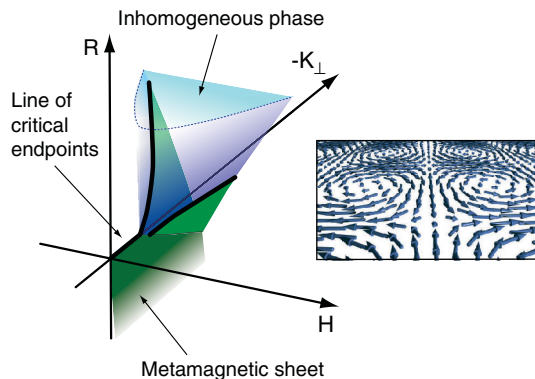


FIG. 3 (color). Phase diagram for the Ginzburg-Landau theory with possible spin texture. Green sheets represent first-order transitions in ϕ . Blue sheets represent continuous transitions into the inhomogeneous phase. The possible spin texture is constructed from four spin helices arranged in a square.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

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 four-fold lattice symmetry (as in $\text{Sr}_3\text{Ru}_2\text{O}_7$) suggests four preferred wave vectors. There are two ways to superpose these: in pairs of $\pm\mathbf{q}$ 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 , θ 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 $\text{Sr}_3\text{Ru}_2\text{O}_7$ 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

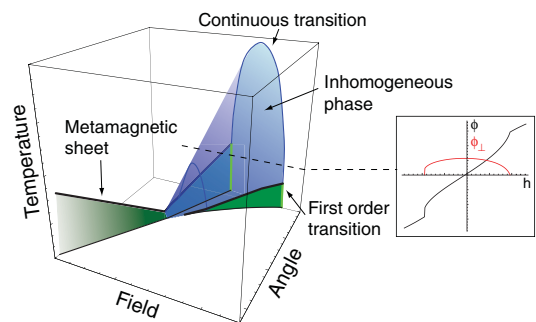


FIG. 4 (color). The phase diagram rotated into the experimental orientation. The dashed line shows a trajectory through the inhomogeneous region. The inset shows the variation of longitudinal and transverse magnetization through this trajectory.

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, pseudoelastic 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 d -wave distortion of the Fermi surface [33]. The topology of the resulting phase should be similar to ours if extended in angle. The main distinction is in the spatial modulation that we predict, which could be probed directly by neutron scattering.

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., NbFe_2 [34] exhibits a peak in resistivity associated with the bifurcation of a metamagnetic transition and finite wave vector magnetic order and ZrZn_2 [35] may show similar features.

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