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Yasuyuki Kato

Los Alamos National Laboratory Theoretical Division

C. D. Batista

Los Alamos National Laboratory Theoretical Division

I. Vekhter

Louisiana State University

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Structure of magnetic order in Pauli limited unconventional superconductors

Yasuyuki Kato¹, C. D. Batista¹, I. Vekhter²

¹*Theoretical Division, T-4 and CNLS, Los Alamos National Laboratory, Los Alamos, NM 87545 and*

²*Department of Physics and Astronomy, Louisiana State University, Baton Rouge, Louisiana, 70803, USA*

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We analyse the magnetic structure of the antiferromagnetic order induced in Pauli limited d -wave superconductors by Zeeman coupling to magnetic field. We determine the phase diagram in the H - T plane, and find that the magnetic phase, which is stabilized at low temperatures and just below the upper critical field, can have two realizations depending primarily on the shape of the underlying Fermi surface. The double- \mathbf{Q} magnetic ordering may persist over the entire coexistence range. Alternatively, there may exist a weak first order transition from a double- \mathbf{Q} structure at lower fields to a single- \mathbf{Q} modulation at higher fields. Together with the calculations of the NMR line-shape these results suggest the second scenario as a serious candidate for describing the superconducting state of CeCoIn₅.

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I. INTRODUCTION.

Understanding of the emergence and stability of competing orders in correlated systems has been a major focus of research. Static long-range order changes the excitation spectrum of itinerant systems, and the resulting energy gain determines which instability dominates. A single order parameter appears in simple cases, but electron-electron interactions can also favor several distinct phases in many materials. The competition between different ordering phenomena make these phases sensitive to applied pressure, chemical doping and magnetic field.

Heavy fermion CeCoIn₅ presents one of the most prominent and puzzling examples of such complex behavior. At ambient pressure it is a very clean singlet d -wave superconductor with lines of nodes in the gap function. Upon doping, superconductivity coexists with and then is pre-empted by an antiferromagnetic (AFM) order. The isostructural CeRhIn₅ is an AFM metal. At low temperatures and fields, H , just below the Pauli limited upper critical field H_{c2} , CeCoIn₅ enters a thermodynamic phase that was initially conjectured¹ to be the first realization of the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state, where the superconducting (SC) order parameter oscillates in real space with a length scale proportional to the ratio between H and the Fermi velocity. However, experiments showed that superconductivity coexists with static long-range incommensurate AFM order in this phase²⁻⁷, even though no AFM order is found upon suppression of SC by doping, magnetic field, or pressure⁸⁻¹¹. Incommensurability and small ordered moment are consistent with itinerant, or spin-density wave (SDW) magnetism, which normally competes for the electronic states with superconductivity. The inescapable conclusion that in CeCoIn₅ superconductivity enables antiferromagnetism challenged our views.

The existing theories fall broadly into two categories. The first assumes that the spatial variation of the SC order under applied field (due to the FFLO modulation) yields a higher single-particle density of states (DOS)

in the regions where superconductivity is suppressed, and nucleates a SDW¹²⁻¹⁵. However, single NMR line shape indicates homogeneous magnetism^{5,16}, and, together with independence of the SDW wave vector on the field⁶, suggests that the origin of the SDW instability is not related to this modulation (although the FFLO state may still exist¹⁶). The second category investigates how a strong Pauli limiting in a nodal superconductor may promote a uniform SDW state¹⁷⁻²¹. We recently showed²¹ that a two-dimensional d -wave superconductor under a Zeeman (paramagnetic) field is *generically* unstable towards formation of a SDW at the wave vectors $\pm\mathbf{Q}_{\pm}$ connecting the opposite nodes of the SC order parameter. The instability is due to nearly perfect nesting of field-induced pockets of Bogoliubov quasiparticles, and explains the incommensurate and field-independent SDW wave vector, as well as the direction of the local moment normal both to the applied field and to the structural layers.

The main challenge to these theories is the structure of the SDW state. Very generally, if the instability originates from magnetic scattering of quasiparticles between the nodal regions, the staggered magnetization, $m_{\mathbf{Q}}$, of a d -wave superconductor has Fourier components for the four wave vectors $\pm\mathbf{Q}_{\pm}$ connecting the “nested” pairs of nodal points, i.e. along each of the two orthogonal directions, 110 and $1\bar{1}0$. We term this structure double- \mathbf{Q} (2Q). The 2Q SDW gaps all four pockets of Bogoliubov quasiparticles thus leading to the greatest energy gain. Therefore, it is always more stable for weak-coupling and hence lower fields²¹. However, the NMR lineshape^{5,16} is consistent solely with a single- \mathbf{Q} (1Q) SDW modulation. We conjectured previously that the 1Q state may be stabilized by moving away from the weak-coupling limit, but, to our knowledge, no resolution of this discrepancy, which is generic to the theories of SDW instability of the nodal superconductors, has been offered.

Below we show that interference between Bogoliubov quasiparticles from the neighborhood of different pockets is the relevant factor that determines the magnetic

structure. The phase diagram contains a transition line between 2Q and 1Q magnetic orders for a range of parameters away from the weak-coupling limit. This transition has weak thermodynamic signatures (such as the specific heat anomaly) but manifests itself very clearly in the change of the NMR lineshape. Unambiguous observation of such a change would strongly favor the current theory for the emergence of the SDW order in CeCoIn₅.

II. MAGNETIC STRUCTURE OF THE COEXISTENCE PHASE

We consider a mean-field Hamiltonian for a d -wave superconductor under Zeeman magnetic field,

$$\begin{aligned} \mathcal{H} &= \mathcal{H}_{\text{BCS}} + \mathcal{H}_{\text{M}} + \frac{N|\Delta_0|^2}{V} + 2NJ(m_{\mathbf{Q}_+}^2 + m_{\mathbf{Q}_-}^2), \\ \mathcal{H}_{\text{BCS}} &= \sum_{\mathbf{k}, \sigma} (\epsilon_{\mathbf{k}} - h\sigma) c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} - \sum_{\mathbf{k}} (\Delta_{\mathbf{k}} c_{\mathbf{k}\uparrow}^\dagger c_{-\mathbf{k}\downarrow}^\dagger + \text{H.c.}) \\ \mathcal{H}_{\text{M}} &= -J \sum_{\mathbf{k}, \nu=\{\pm\}} (m_{\mathbf{Q}_\nu} c_{\mathbf{k}-\mathbf{Q}_\nu\uparrow}^\dagger c_{\mathbf{k}\downarrow} + m_{\bar{\mathbf{Q}}_\nu} c_{\mathbf{k}+\mathbf{Q}_\nu\uparrow}^\dagger c_{\mathbf{k}\downarrow} + \text{H.c.}). \end{aligned}$$

Here $\epsilon_{\mathbf{k}} = 2t(\cos k_x + \cos k_y) + 4t' \cos k_x \cos k_y - \mu$ is the band energy, $h = g\mu_B H/2$ is the Zeeman splitting, N is the number of lattice sites, and $\Delta_{\mathbf{k}} = \Delta_0(\cos k_x - \cos k_y)$ is the superconducting order parameter. J denotes magnetic interaction, and we explicitly separated the four (equal in magnitude) wave-vectors $\pm\mathbf{Q}_\nu$: $\nu = \pm$ indicates a different pair of "nested" gap nodes, while \mathbf{Q}_ν and $\bar{\mathbf{Q}}_\nu = -\mathbf{Q}_\nu$ are the two vectors connecting a given pair, see inset of Fig.1(a). Below we select $\mathbf{Q}_\pm = (7\pi/6, \pm 7\pi/6)$ by fixing the chemical potential to $\mu = \epsilon_{\mathbf{Q}_+/2}$. The self-consistency equations are

$$\Delta_0 = \frac{V}{N} \sum_{\mathbf{k}} (\cos k_x - \cos k_y) \langle c_{\mathbf{k}\uparrow}^\dagger c_{-\mathbf{k}\downarrow}^\dagger \rangle, \quad (1)$$

$$m_{\mathbf{Q}} = \frac{1}{N} \sum_{\mathbf{k}} \langle c_{\mathbf{k}+\mathbf{Q}\uparrow}^\dagger c_{\mathbf{k}\downarrow} \rangle, \quad (2)$$

where $\langle \dots \rangle$ denotes the thermodynamic average for \mathcal{H} , and V is the pairing interaction in the d -wave channel.

The four order parameter components $m_{\mathbf{Q}}$ lead to the following real space distribution of magnetic moments

$$\begin{aligned} m_{\mathbf{r}}^x &= \frac{1}{2} \sum_{\nu=\{\pm\}} \left[m_{\mathbf{Q}_\nu} e^{i\mathbf{Q}_\nu \cdot \mathbf{r}} + m_{\bar{\mathbf{Q}}_\nu} e^{-i\mathbf{Q}_\nu \cdot \mathbf{r}} \right], \\ m_{\mathbf{r}}^y &= \frac{i}{2} \sum_{\nu=\{\pm\}} \left[m_{\mathbf{Q}_\nu} e^{i\mathbf{Q}_\nu \cdot \mathbf{r}} - m_{\bar{\mathbf{Q}}_\nu} e^{-i\mathbf{Q}_\nu \cdot \mathbf{r}} \right]. \end{aligned} \quad (3)$$

The easy axis in CeCoIn₅ is perpendicular to the layers²², so the field in the plane clearly favors the staggered moment along the easy direction, in agreement with experiment^{22,23}. We therefore assume such collinear spin ordering, $|m_{\mathbf{Q}_\nu}| = |m_{\bar{\mathbf{Q}}_\nu}|$, and choose the easy axis as the spin x -direction (crystal c -axis), $m_{\mathbf{Q}_\nu} = m_{\bar{\mathbf{Q}}_\nu}^*$. There is a

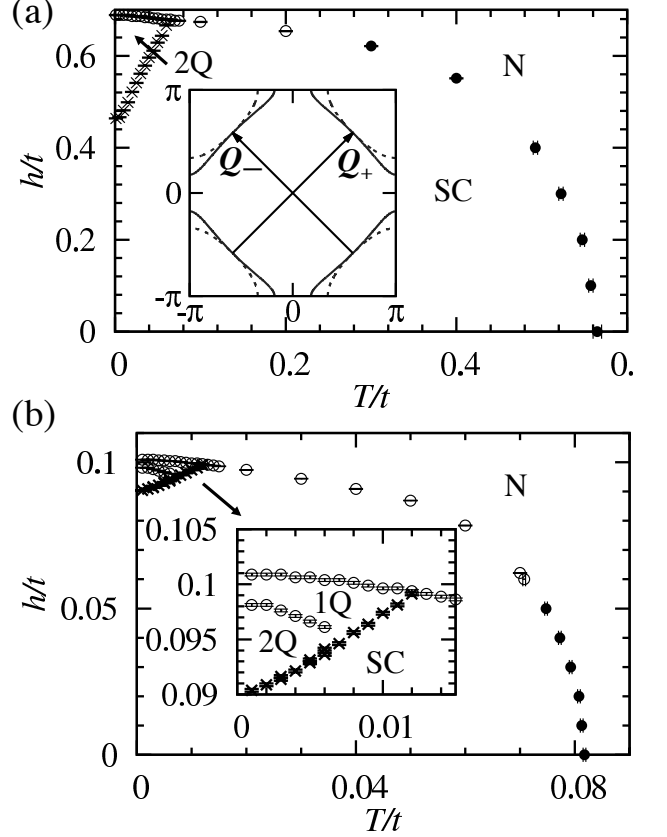


FIG. 1: Thermodynamic phase diagrams for (a) $t'/t = 0$, $V/t = 3$, $J/t = 4$, and (b) $t'/t = 0.2$, $V/t = 0.8$, $J/t = 2.4$. Open and closed circles represent first and second order phase transition, respectively. Crosses represent the transition from single-Q or double-Q to uniform superconductivity for $L = 432$. The inset of (a) shows the nesting vectors \mathbf{Q}_\pm and the Fermi surfaces at $t'/t = 0$ (dashed line) and $t'/t = 0.2$ (solid line), with $V/t = 0$, and $J/t = 0$.

zero phase mode for each value of ν due to the incommensurate nature of the ordered phase. Without loss of generality we choose the amplitudes $m_{\mathbf{Q}}$ to be real numbers, i.e., we fix both phases at zero. Two possible magnetic structures can be stabilized under these conditions. The 1Q structure, in which only one of the two amplitudes, $m_{\mathbf{Q}_\nu}$, is finite, corresponds to a SDW that is modulated along the direction parallel to the corresponding \mathbf{Q}_ν vector. The 2Q structure, $|m_{\mathbf{Q}_+}| = |m_{\mathbf{Q}_-}|$, is modulated along both 110 and $1\bar{1}0$.

We solved Eqs. (1)-(2) on a finite square lattice of linear sizes up to $L = 864$ and for different values of band parameters and coupling constants. Above we chose the set of four incommensurate ordering wave vectors, $(\pm\mathbf{Q}, \pm\mathbf{Q})$, of the form $\mathbf{Q} = (n+1)\pi/n$ with $n = 6$. With this choice the number of sites in the new unit cell is $2n^2$. Hence to solve the mean field Hamiltonian \mathcal{H} , we need to diagonalize a matrix of size $8n^2$ at each \mathbf{k} -point of the original Brillouin Zone (the additional factor of four arises from the spin index and from the anomalous terms

that create or annihilated pairs with opposite momenta). For our choice of $n = 6$, we diagonalized matrices of dimension 288×288 and then summed over momentum space to compute the free energy density:

$$f_{MF} = -\frac{k_B T}{L^2} \ln [\text{Tr} e^{-\beta \mathcal{H}}]. \quad (4)$$

We found two distinct types of phase diagrams that are shown in Figs. 2. Both of them show the 2Q ordering at low temperatures and moderately high fields. However, while this phase persists throughout the whole magnetic region for some parameter values, as is shown in Fig. 1(a), in other cases the SDW phase exhibits a transition between 1Q and 2Q structures [see Fig. 1(b)]. The transition between uniform SC and normal (N) phases is the same in both situations: second order at low fields/high temperatures, and first order at high fields/low temperatures; this behavior is generic for two-dimensional paramagnetically limited superconductors. The transition from the normal to the magnetically ordered SC phase remains of first order, while the onset of SDW in the SC phase occurs via a second order transition. Finally, a weak first order phase transition line separates the 1Q and 2Q phases. Since neutron scattering²³ and NMR measurements⁵ of CeCoIn₅ clearly indicate the presence of a 1Q phase at high fields, this latter case is relevant²¹, and we now discuss the origin and the properties of the 1Q/2Q transition in detail.

Recall that in the weak-coupling (low field) regime, the SDW order arises from nesting between opposite pockets of Bogoliubov quasiparticles (eigenstates of $\mathcal{H}_0 + \mathcal{H}_{BCS}$)²¹ generated by the Zeeman term in a nodal superconductor²⁴. The stability of the 2Q phase arises from the decoupling of the mean field equations (2) for the four amplitudes $m_{\mathbf{Q}}$, which requires the same amplitude $|m_{\mathbf{Q}}|$ of all the components to fully gap the four nodal pockets. Numerical solutions for all sets of parameters always yield the 2Q order at the lowest fields, in agreement with this argument.

The lowest order interaction term of a Ginzburg-Landau expansion of the free energy is $|m_{\mathbf{Q}_+}|^2 |m_{\mathbf{Q}_-}|^2$. Diagrammatically, the coefficient of this term involves a product of four propagators of the Bogoliubov quasiparticles at momenta separated by $\pm \mathbf{Q}_{\pm}$, with the main contribution when all four momenta are near the Fermi surface pockets. Since the ordering wave vectors \mathbf{Q}_{\pm} are incommensurate, the near-nodal Bogoliubov quasiparticles do not satisfy this requirement and give a small contribution to this coefficient. In contrast, once the pockets grow to include areas away from the nodes, near $(\pm\pi, 0)$ and $(0, \pm\pi)$ points, the constraint is satisfied. Therefore the coefficient of the $|m_{\mathbf{Q}_+}|^2 |m_{\mathbf{Q}_-}|^2$ is small at low fields, but, as the field increases and the pockets grow, the two components of the magnetization begin to interfere destructively, leading to an increase in the energy of the 2Q phase relative to that of the 1Q phase. To test this argument we computed the average occupation number of Bogoliubov quasiparticles with a given wave-vector \mathbf{k}

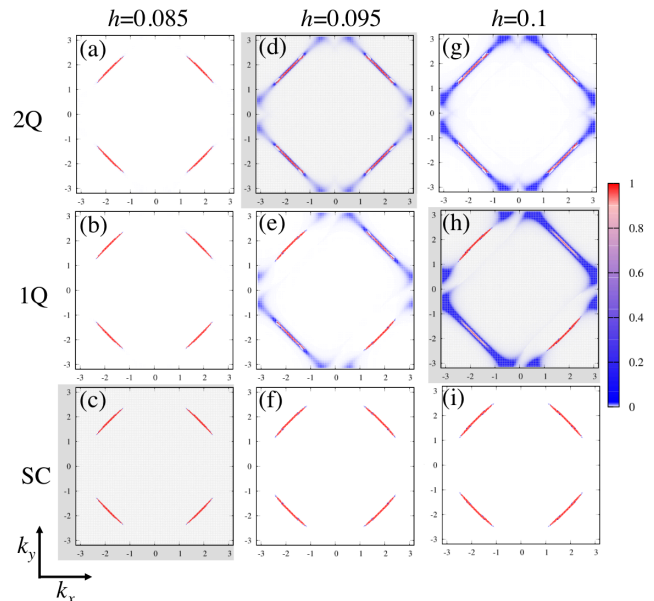


FIG. 2: Density of Bogoliubov quasi-particles $\langle \gamma_{\mathbf{k}\uparrow}^\dagger \gamma_{\mathbf{k}\uparrow} \rangle$ for $t'/t = 0.2$, $V/t = 0.8$, $J/t = 2.4$, and $T/t = 10^{-4}$ with $L = 864$. Shaded panels (c), (d), and (h) correspond to the lowest free energy solution of the self-consistent equations (2) at a given magnetic field value.

over the entire Brillouin zone for the parameters where 2Q/1Q transition exists. The results, shown in Fig. 2, clearly indicate that the 1Q phase becomes stable as soon as the overlap between particle-hole clouds from different pockets becomes strong. Therefore, the main physical parameter that decides between the two possible scenarios shown in Figs. 1(a) and (b) is the barrier between orthogonal pockets which, in turn, depends on the shape of the Fermi surface. Recall that the SDW phase appears for values of the magnetic interaction $J \geq 0.85 - 0.9J_c$, where J_c is the critical value when the normal state becomes unstable towards magnetic order²¹; this result remains valid for any Fermi surface shape. We verified that, for a Fermi surface where 2Q-1Q transition exists, the 1Q phase persists over a wide range of values of the pairing interaction V , although once V is significantly reduced, the 1Q region becomes too narrow to be found numerically in our calculation. At the same time, for the Fermi surface where only 2Q order is found, such as that used in the main panel of Fig. 1(a), no changes in the interaction can induce the 1Q phase.

Given that the 1Q and 2Q phases can only exist together in the phase diagram, we analyse the phase transition between the two. Figures 3(a-c) show the solutions of Eqs.(1)-(2) at low T for the parameters in Fig. 1(b). While both the SC and the SDW order parameters show a significant jump at the transition from the normal paramagnetic phase, the change in Δ_0 at the 1Q-2Q transition is extremely small, and the reduction in $m_{\mathbf{Q}_+}$ is compensated by the concomitant in-

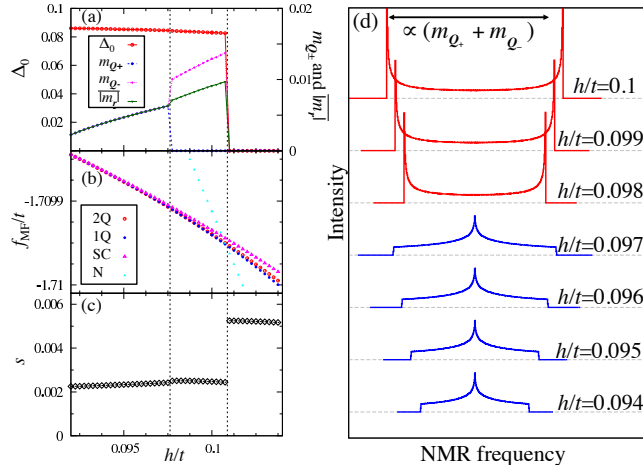


FIG. 3: Mean-field solutions for $t'/t = 0.2$, $V/t = 0.8$, $J/t = 2.4$, and $T/t = 0.003$ with $L = 432$. (a) Order parameters (Δ_0 , m_{Q+} , and m_{Q-}) and the spatially-averaged magnetization $\overline{|m_r|}$. (b) Free energy densities f_{MF}/t for different solutions of self-consistent equations (2). (c) Entropy s (computed by taking the derivative of the free energy density as a function of temperature) as a function of magnetic field. (d) Expected shape of the NMR spectrum, see text for details.

crease in m_{Q-} , so that the spatially-averaged magnetization $\overline{|m_r|}^2 \equiv (1/L^d) \sum_r m_r^2 = (m_{Q+}^2 + m_{Q-}^2)/2$, remains nearly unchanged. This is also clear from the almost unnoticeable kink in the free energy density at the transition line. As a result, the latent heat at the transition between N and 1Q phases is much larger than the latent heat at the 1Q-2Q transition (see Fig. 3(c)). In other words, the 1Q-2Q transition, while first order, has very weak thermodynamic signatures, and is difficult to detect by bulk measurements. On the other hand, probes sensitive to the local magnetic structure, such as neutron scattering or NMR, should be able to clearly distinguish these two different phases. Figure 3(d) shows the predicted qualitative change of the NMR line shape across the transition in the magnetic structure of the Q -SDW phase and for different values of the magnetic field. We plot the density

distribution of the local magnetization for incommensurate ordering, and are not including the details of the dipolar and hyperfine interactions that would be needed for a quantitatively accurate determination of the NMR line shape. Our main goal is to demonstrate the qualitative difference between the signals in the 1Q and the 2Q phases: while the 1Q phase leads to a double-horn NMR line shape characteristic of unidirectional modulation, the profile in the 2Q phase has a maximum at the center (the logarithmic Van-Hove singularity). The NMR line shape as a function of increasing field, see Fig. 3(d), is very consistent with recent measurements from two different groups^{5,16}.

III. CONCLUSIONS

In summary, we presented a theory for the magnetic structure of the low-temperature and high field phase of Pauli limited nodal superconductors such as CeCoIn₅. Our main finding is that such materials are expected to broadly fall into two classes depending predominantly on the shape of the underlying Fermi surface: those exhibiting only the phase in which the local moments are modulated along the two orthogonal directions connecting opposite nodes of the superconducting gap, and those exhibiting a transition between such a doubly modulated phase and SDW order along a single inter-nodal direction. The origin of the transition is in the destructive interference of scattering processes of Bogoliubov quasiparticles with two orthogonal wave-vectors. Thermodynamic signatures of the 2Q-1Q transition are very weak, and therefore local magnetic probes are best suited for probing it. The NMR line shape in our analysis is in a good agreement with recent data. It would be highly desirable to further test the proposed transition using neutron scattering.

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