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We report the discovery of incommensurate magnetism near quantum criticality in CeNiAsO through neutron scattering and zero field muon spin rotation. For $T < T_{x0} = 8.7(3)$ K, a second order phase transition yields an incommensurate spin density wave with wave vector $k = (0.44(4), 0.0)$. For $T < T_{x2} = 7.6(3)$ K, we find co-planar commensurate order with a moment of $0.37(5) \mu_B$, reduced to 30% of the saturation moment of the $|\pm \frac{1}{2}\rangle$ Kramers doublet ground state, which we establish through inelastic neutron scattering. Muon spin rotation in CeNiAs$_{1-x}$P$_x$O shows the commensurate order only exists for $x \leq 0.1$ so we infer the transition at $x_c = 0.4(1)$ is between an incommensurate longitudinal spin density wave and a paramagnetic Fermi liquid.

The competing effects of intra-site Kondo screening and inter-site Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions in rare earth intermetallics epitomize the strongly correlated electron problem. While the Néel and Kondo lattice limits are well understood [1], the transition between them is far from. It involves an increase in the volume enclosed by the Fermi surface (FS) as the $4f$ electron is incorporated on the Kondo lattice side of the transition[2, 3]. Deviations from the $\rho \propto T^2$ dependence of resistivity is interpreted as indicative of the associated quantum criticality, which is denoted as “local” because it involves the entire FS. In support of this concept, compounds with the requisite transport anomalies have been discovered where physical properties that involve averages over distinct regions of momentum space have related critical exponents. The eventual transition to magnetic order when RKKY interactions dominate can coincide with the localization transition or occur within the large or small FS phases. Clearly the nature of the corresponding quantum critical point is strongly affected as magnetic ordering is momentum selective and breaks time reversal symmetry.

Exploration of model systems is essential to uncover the overall phase diagram of this complex strongly correlated regime. CeCu$_{6-x}$Au$_x$ provided a first example of local criticality. de Haas-van Alpen measurements provide evidence for an abrupt rearrangement of the FS in CeRhIn$_5$ at 2.25 GPa [4–6]. A step change in the Hall coefficient of YbRh$_2$Si$_2$ coupled with anomalous and yet unexplained critical exponents at the field driven ferromagnetic transition have been interpreted as evidence the magnetic and the electron localization transitions coincide[7–11]. Each compound adds unique insights and distinct experimental opportunities.

Isostructural to the 1111 iron pnictides, CeNiAsO is an exciting new addition to the landscape of strongly correlated electron systems [12]. Magnetically ordered at low–T and ambient pressure, substitution of P for As or pressure drives CeNiAs$_{1-x}$P$_x$O to a paramagnetic Fermi-liquid. Non-Fermi-liquid transport is found up to the critical pressure $P_c = 6.5$ kbar and the critical composition $x_c = 0.4(1)$ and a sign change in the Hall coefficient at $P_c$ indicates FS reconstruction [13]. CeNiAsO differs from other systems studied to date in having two magnetic phase transitions [12].

In this letter we determine the corresponding magnetic phases and examine their interplay with FS reconstruction. We show the upper transition is to an incommensurate longitudinal SDW state with wave vector $k = (0.44(4), 0.0)$ that closely matches the umklapp wave vector $(2k_f)$ of the small FS. The second transition yields co-planar commensurate order with a low–$T$ ordered moment reduced to 30% of the saturation moment of the nominal $|\pm \frac{1}{2}\rangle$ Kramers doublet ground state. P doping suppresses the commensurate phase but retains the SDW perhaps all the way to the critical transition.
These patterns can be fitted by magnetic structures that are consistent with the neutron data and a single crystallographic muon stopping site.

We determined the fundamental magnetic wave vector and spin polarization through neutron diffraction. Weak magnetic peaks are apparent at $T = 2$ and $8$ K after subtracting data at $T = 15$ K (Fig. 2 (c-d)). At $T = 2$ K, the difference pattern shows several resolution limited peaks. The peak with the lowest wave vector transfer $Q \approx 0.77 \, \text{Å}^{-1}$ can be accounted for as $Q_m = (0.5, 0, 0)$. Magnetic neutron diffraction probes spin polarization perpendicular to wave vector transfer so this indexing implies spin components along $b$ and/or $c$. Upon warming to $8$ K $< T_{N1}$, the absence of this first peak is indicative of a longitudinal spin density wave (SDW) polarized along $a$. The width of the intensity maxima $T = 8$ K and $Q \approx 1.1 \, \text{Å}^{-1}$ in Fig. 2 (c) exceeds the instrumental $Q$-resolutions. The incommensurability indicated by $\mu$SR can account for this. The magnetic signal at $8$ K is however quite weak and since there is no energy resolution, inelastic magnetic scattering may also contribute to the broadened peaks, particularly near the polarization suppressed $Q_m$ peak. The diffraction data thus do not permit a unique determination of the spin structure for $T_{N2} < T < T_{N1}$. The combination of muon, specific heat, and elastic/inelastic neutron data, however, does allow an accurate determination of both structures.

Using Kovalev notation [20, 21], the reducible magnetic representation associated with $k = (\mu 0 0)$ decomposes into three two-dimensional irreducible representations (IR): $\Gamma_{mag} = 2\Gamma_1^{(2)} + \Gamma_1^{(2)}$ with 6 Basis Vectors (BVs) (Table S2). Landau theory allows only one IR for each of the two second order phase transitions. Below $T_{N1}$, BVs $\psi(4)$ and $\psi(6)$ of $\Gamma_1$ depict a spin structure with...
moments along \( \mathbf{a} \). Adding \( \psi(3) \) and \( \psi(5) \) allows for moments along \( \mathbf{c} \). Below \( T_{N2} \), we can account for the diffraction pattern in Fig. 2 (d) by adding \( \psi(1) \) and \( \psi(2) \) of \( \Gamma_2 \). The best fit corresponds to a reduced \( \chi^2 = 1.95 \) and a staggered moment \( \langle m \rangle = 0.37(5) \mu_B/\text{Ce} \) that is canted by \( \varphi \approx 36(6)^\circ \) to the \( \mathbf{a} \) axis (Fig. 1 (c)). While allowed by symmetry, the diffraction data place a limit of 0.06 \( \mu_B \) on any \( \mathbf{c} \)-component of the staggered moment.

\( \mu\text{SR} \), which probes magnetism in real space, offers an independent assessment of the proposed structures. We find a consistent description of the precession data with the muon stopping site \( \left( \frac{1}{3}, \frac{1}{3}, z_\mu \right) \) in Fig. 1 (a). The fitting analysis described below yields \( z_\mu = 0.1471(3) = z_{\text{Ce}} \), close to the preferred distance of muons from \( \text{O}^{2-} \) [22]. This location is also favored considering the electrostatic potential-energy map for CeFeAsO [23]. The observation of two muon precession frequencies suggests two magnetically inequivalent muon sites (see Fig. 1 (b-c)). The asymmetry pattern \( P_{\mu}^2(t) \) can be fitted to equation S1 wherein the magnetic field distribution function \( \rho_I(B) \) is calculated directly from the spin structures. For the low \( T \) commensurate state, \( \rho_I(B) \) consists of two delta functions corresponding to the magnetic field at each of the two magnetically inequivalent (but crystallographically equivalent) muon sites. The best fit is obtained with moment \( \langle m \rangle = 0.37(2) \mu_B \) and rotation angle \( \varphi = 36(7)^\circ \), which is in excellent agreement with the Rietveld refinement of neutron diffraction. For the high \( T \) incommensurate state, \( \rho_I(B) \) is continuous: The incommensurate nature of the spin structure ensures every muon site, though crystallographically equivalent, is magnetically unique and contributes a distinct precession frequency. The best fit leads to an incommensurate wave vector \( \mathbf{k} = (0.44(4), 0.0), m_c = 0.27(6) \mu_B, \) and \( m_a = 0.08(3) \mu_B \).

The corresponding calculated muon asymmetry and neutron diffraction are in Fig. 2 (a) \& (c). A small component of \( m_c \) implies this is a magnetic cycloid. The corresponding lack of inversion symmetry could have interesting consequences for electronic transport. However, since \( m_c \ll m_a \) we retain the terminology of a longitudinal SDW. In summary, the spin structures for two ordered states -- a longitudinal SDW (Fig. 1 (b), Fig. S4) and a commensurate coplanar structure (Fig. 1 (c)) -- account for both neutron and \( \mu\text{SR} \) data.

For context we examine the \( 4f \) electron crystal field excitations through inelastic magnetic neutron scattering (Fig. 4 (a-e)). At \( T = 7 \) K, the intensities of modes at \( E \approx 10 \) meV, 30 meV and 40 meV rise with \( Q^2 \) and are observed both for CeNiAsO and non-magnetic LaNiAsO and so must be vibrational [27]. In the difference data \( \tilde{I}(Q, E) \) and \( \tilde{I}(E) \) (Fig. 4 (c-e)), we associate the two broad modes at \( E_1 \approx 18(3) \) meV and \( E_2 \approx 70(8) \) meV with magnetic excitations because their intensity decreases with \( Q \) as the \( 4f \) formfactor. In the tetragonal environment of Ce\(^{3+} \), the \( J = \frac{5}{2} \) multiplet splits into three Kramer’s doublets. The two magnetic modes are correspondingly assigned to crystal-field-like excitations from the ground state (GS) to two excited doublets. At \( T = 200 \) K population of the excited state yields a broad mode at 50 meV \( \approx E_2 - E_1 \), which arises from excitations between the excited doublets. Finally we observe a sharp mode at \( E_0 \approx 2 \) meV within the AFM ordered state (inset, Fig. 4 (d)). In the language of CEF theory, this is an intra-doublet transition driven to inelasticity by the molecular exchange field. As expected for a strongly correlated solid, the crystal field excitations measured for a powder sample are broadened by damping and
FIG. 3: Temperature dependence of (a) the longitudinal \(m_a\) and (b) the transverse moments \(m_b\) for high \(T\) and \(m_c\) for low \(T\) phase. Black dots were extracted from Rietveld fits to neutron diffraction data. The 2 K and 8 K data points were averaged over two chopper settings. Blue diamonds were inferred from \(\mu\)SR fits. The solid lines are guides to the eye. (c) Temperature dependence of the averaged static field. (d) Specific heat \(C_p/T\) in zero field and for \(\mu_0H = 14\) T. The upturn in \(C_p/T\) at 14 T is due to the nuclear spin contributions as indicated by the solid red line.

dispersion, leading to the half width at half maximum (HWHM) of \(\Gamma_1 = 13\) meV and \(\Gamma_3 = 24\) meV. Fitting to Lorentzian spectral functions leads to HWHM of \(\Gamma_0 = 2\) meV that is comparable to the Kondo temperature \(T_K = 15(5)\) K inferred from thermo-magnetic data [12].

Given these broad modes, a local moment crystal field model cannot be comprehensive but it provides a useful starting point. As detailed in the SI, we carried out a global fit of a symmetry-constrained crystal field model to the normalized scattering data \(\tilde{I}(E)\) at \(T = 7\) K and 200 K. After optimizing the crystal field parameters a molecular exchange field and three transition specific relaxation rates, Fig. 4(d-e) shows a consistent description of data from two instrumental configurations and two temperatures is achieved. The model also accounts for the temperature dependent susceptibility data. Consistent with the easy plane (ab plane) character of the ordered states, the GS wave function is \(|\pm \frac{1}{2}\rangle\) (\(\Gamma_7\)).

As indicated in the DFT FS plot (Fig. 1 (d)), the ordering wave vector \(k = (0.44(4), 0, 0)\) satisfies a nesting condition. This suggests the ordered state for \(T_{N2} < T < T_{N1}\) should be classified as a SDW [28–32]. It is common for incommensurate (IC) magnets to undergo a longitudinal to transverse spin reorientation transition that reduces the modulation in the magnitude of the dipole moment per unit cell while sustaining the IC modulation [30, 33]. The situation is different for CeNiAsO, which not only develops transverse magnetization but also becomes commensurate for \(T < T_{N2}\). To arrive at the spin structure in Fig. 1 (c) from the commensurate version of Fig. 1 (b) involves counter-rotating the upper and lower AFM layers of a CeO sandwich (Fig. 1 (a)) by \(\varphi = 36^\circ(5)\) around c. While inter-layer bi-linear interactions vanish at the mean field level for \(k = (0.5, 0, 0)\) type order, inter-layer bi-quadratic interactions[34, 35] give rise to a term in the free energy of the form \((m^2 \cos 2\varphi)^2\) that can favor \(\varphi = 45^\circ\) for a commensurate structure only. As \(m\) grows upon cooling this term can be expected to induce both the IC to commensurate transition and the symmetry breaking transverse magnetization at \(T_{N2}\).

This brings us to the character of magnetism in CeNiAs\(_{1-x}\)P\(_x\)O. Upon cooling, CeNiAsO passes from Fermi liquid to IC SDW to commensurate non-collinear order in two second order phase transitions. P doped samples that we examined (CeNiAs\(_{1-x}\)P\(_x\)O for \(x > 0.1\))
all show the characteristic $\mu$SR oscillation associated with IC magnetism (Fig.2 (a)) down to 50 mK. This indicates the commensurate state is limited to a low $T$, low $x$ pocket (Fig. 1 (d)) and the initial instability of the strongly correlated Fermi liquid in CeNiAs$_{1-x}$P$_x$O is to an IC SDW. An important open question is whether the characteristic wave vector of the SDW evolves with $x$ or continues to be associated with the small FS as for $x = 0$.

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[48] See Supplementary Material for the following information: the sample synthesis [36], details in the Rietveld refinement [37, 38], details in the DFT calculation [39–41], analysis of Schottky anomaly [42–44] in the low-T specific heat, crystal field analysis and corresponding magnetic susceptibility calculation [45–47].