Distinct itinerant spin-density waves and local-moment antiferromagnetism in an intermetallic ErPd$_2$Si$_2$ single crystal

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Identifying the nature of magnetism, itinerant or localized, remains a major challenge in condensed-matter science. Purely localized moments appear only in magnetic insulators, whereas itinerant moments more or less co-exist with localized moments in metallic compounds such as the doped-cuprate or the iron-based superconductors, hampering a thorough understanding of the role of magnetism in phenomena like superconductivity or magnetoresistance. Here we distinguish two antiferromagnetic modulations with respective propagation wave vectors at $Q_x = (H \pm 0.557(1), L \pm 0.150(1))$ and $Q_C = (H \pm 0.564(1), 0, L)$, where $(H, L)$ are allowed Miller indices, in an ErPd$_2$Si$_2$ single crystal by neutron scattering and establish their respective temperature- and field-dependent phase diagrams. The modulations can co-exist but also compete depending on temperature or applied field strength. They couple differently with the underlying lattice albeit with associated moments in a common direction. The $Q_x$ modulation may be attributed to localized 4f moments while the $Q_C$ correlates well with itinerant conduction bands, supported by our transport studies. Hence, ErPd$_2$Si$_2$ represents a new model compound that displays clearly-separated itinerant and localized moments, substantiating early theoretical predictions and providing a unique platform allowing the study of itinerant electron behavior in a localized antiferromagnetic matrix.

Unravelling the interplay between opposite but also complementary phenomena stands at the forefront of condensed-matter science. For example, unconventional Cooper pairs, on the one hand, can be glued by the common thread of spin fluctuations; on the other hand, they can be competitively ruined by the formation of long-ranged ferromagnetic (FM) or antiferromagnetic (AFM) ordering. The archetypal picture of magnetism displays an opposing dual character, i.e., itinerant electron magnetism with weak interactions and localized moments with strong Coulomb repulsions. Understanding the behaviour of itinerant electrons in the presence of localized moments may shed light on nontrivial properties of correlated electron systems such as spin-or charge-density waves or superconductivity, for which unifying both types of magnetism in one compound with clear electronic origins is necessary. Experimentally, it is hard to clearly distinguish itinerant from localized moments in 3d-based strongly-correlated electron materials like the copper-oxygen superconductors, despite the fact that they can be theoretically modeled. Consequently, the experimentally observed smaller moment size in point compared to the expected theoretical saturation value can be attributed either to the screening effect of itinerant electrons or to the frustration effect of localized spins. In addition, the nature of the antiferromagnetism or spin-density waves (SDWs) of iron-superconductors is still hotly debated as to whether the magnetic neutron excitations can be best described by the itinerant or localized picture. Disentangling these arguments necessitates the search for a model system that hosts clearly-defined itinerant and localized spins, thus permitting a complete understanding of their coupling mechanism.

The 5f electrons in actinides such as U-based compounds UPt$_3$ and UPd$_2$Al$_3$ show experimental and theoretical evidence for a localized and delocalized dual nature which may play an important role in producing...
heavy-fermion superconductivity\textsuperscript{46}. In strongly spin-orbit coupled systems such as 4f - or 5f-based compounds, a novel spin-orbit density wave was proposed as an emergent quantum phase with a breaking of translational while preserving time-reversal symmetry, which theoretically sheds light on the intriguing “hidden-order” of URu\textsubscript{2}Si\textsubscript{2}\textsuperscript{39}. For lanthanide-based compounds, the rare-earth (RE, except cerium) 4f moments are generally localized because unpaired 4f electrons are well shielded by the 5s\textsuperscript{p} shells. The 4f\textsuperscript{5}d\textsuperscript{6}s\textsuperscript{2} (n = 2–7 and 9–14; m = 0, 1) valence electrons in lanthanide-based conductors act usually as a mediator for the interactions between 4f moments in Ruderman-Kittel-Kasuya-Yosida (RKKY) exchanges (i.e., the long-range ordered 4f moments interact indirectly with each other via conduction bands since the direct coupling between localized 4f moments is generally weak)\textsuperscript{40}, or are ferromagnetically polarized into magnetic polarons (i.e., local short-range FM regimes) by the localized 4f moments\textsuperscript{41-47}. It is thus difficult for the conduction electrons to form a long-ranged AFM ordering. However, taking into account the coupling between nesting electrons and hole parts of Fermi surface, the 4f-based conductors could provide the possibility for combining localized 4f moments and SDWs of itinerant electrons.

The SDW state, a low-energy self-organized collective modulation of electron spins, often appears in electronic conducting materials such as organic linear-chain compounds, low-dimensional metals or superconductors\textsuperscript{13,23-25,27}. Since their first observation in chromium\textsuperscript{23}, SDWs display progressively appealing low-temperature properties, e.g., a proximity with charge-density waves (CDWs) and unconventional superconductivity\textsuperscript{13,22,25,27,44}. Above a threshold field, SDWs can be described as a set of delocalized AFM spins\textsuperscript{25}. Intermetallic REPd\textsubscript{2}Si\textsubscript{2} silicides all crystallize with the same ThCr\textsubscript{2}Si\textsubscript{2}-type (Fig. 1) tetragonal I4/mmm structure \textit{(a = b = 4.0987(1) Å, and c = 9.8762(1) Å at ambient conditions, as listed in Table 1) as that of the family of 122-iron-pnictides\textsuperscript{45-47} and exhibit a wide range of interesting physical properties, e.g., pressure-induced superconductivity in CePd\textsubscript{2}Si\textsubscript{2} and anomalous valence fluctuations in EuPd\textsubscript{2}Si\textsubscript{2}\textsuperscript{45}. Early theoretical proposals\textsuperscript{49} for the 4f conductors with extended RKKY-interactions predicted that localized 4f moments can promote a SDW state in the itinerant conduction electrons. So far, to our knowledge, no clear example of such a material with distinguishable propagation vectors has been identified. Here we report on the first single-crystal neutron scattering study of ErPd\textsubscript{2}Si\textsubscript{2}\textsuperscript{50-55}. We discover two distinct incommensurate spin states and attribute one to the localized 4f electrons while attributing the other mainly to itinerant valence bands. We also build a detailed knowledge of the virtual coupling between both states, which is actually intractable in 3d-metallic systems. Our findings correspond to theoretical predictions\textsuperscript{49} and thus establish a new model material.

Results

Neutron diffraction with polarization analysis. Figure 2 shows data from neutron diffraction with polarization analysis after the sample was cooled to 3 K in zero field. The non-spin-flip (NSF) channel shows only the nuclear Bragg reflections with two incommensurable propagation vectors. One set of peaks, that will be called \textit{Ic} and \textit{Ib}. (Fig. 3), appear at \textit{Qc} = (H ± 0.557(1), 0, L ± 0.150(1)) and the second set, which will be called \textit{Ic}. (Fig. 3), appear at \textit{Qc} = (H ± 0.564(1), 0, L), where (H, L) are the Miller indices for allowed nuclear reflections. Nuclear coherent scattering of polarized neutrons will not flip the neutron spin. Magnetic scattering via polarized neutrons can flip the neutron spin and is determined by the relative direction of the neutron polarization vector \textbf{P} with regard to both the scattering vector \textbf{Q} and the direction of the ordered-moments \textbf{\mu}. In our study, \textit{P} is normal to the scattering plane (H, 0, L), i.e., parallel to the \textit{b} (or \textit{a}) axis. In this case, the moment-dependent cross-sections can be written as:

\[
\left(\frac{d\sigma}{d\Omega}\right)_{SF} = \frac{2}{3} \left(\frac{d\sigma}{d\Omega}\right)_{nsi} + \left(\frac{d\sigma}{d\Omega}\right)_{mag},
\]

\[
\left(\frac{d\sigma}{d\Omega}\right)_{NSF} = \frac{1}{3} \left(\frac{d\sigma}{d\Omega}\right)_{nsi} + \left(\frac{d\sigma}{d\Omega}\right)_{nuc} + \left(\frac{d\sigma}{d\Omega}\right)_{mag},
\]

where the subscripts NSF and SF refer to the non-spin flip and spin flip cross-sections, respectively, \textit{nsi} means the nuclear spin incoherent contribution, and \textit{nuc} refers to nuclear coherent and isotopic

**Table 1 | Summary of the refined room-temperature structural parameters of single-crystal ErPd\textsubscript{2}Si\textsubscript{2} from X-ray powder diffraction**

| Structure | Tetragonal [ThCr\textsubscript{2}Si\textsubscript{2}]-type, I4/mmm |
|-----------|----------------------------------|
| ErPd\textsubscript{2}Si\textsubscript{2} | 4.0987(1), 90 | 9.8762(1), 90 |
| Atom | Site | \textit{x} | \textit{y} | \textit{z} | \textit{B} (Å\textsuperscript{2}) |
| Er | 2\textit{a} | 0 | 0 | 0 | 8 (Å\textsuperscript{2}) |
| Pd | 4\textit{d} | 0 | 0.5 | 0.25 | 3.53(3) |
| Si | 4\textit{e} | 0 | 0 | 0.3798(2) | 3.62(6) |
| R\text{~p} (%) | 3.09; R\text{~wp} (%) | 4.41; R\text{~p} (%) | 2.70; R\text{~p} (%) | 2.19; \text{R}^2 | 3.20. |
The magnetic contributions, labelled by $\text{mag}$, are from two parts: the component of $\mu$ that is along the direction of $\vec{P}$ contributes to NSF scattering, i.e.,

$$
\frac{d\sigma}{d\Omega} \bigg|_{\text{mag}} \propto \langle \mu \parallel \vec{P} \rangle^2;
$$

and the component that is normal to both $\vec{P}$ and $\vec{Q}$ gives rise to SF scattering, i.e.,

$$
\frac{d\sigma}{d\Omega} \bigg|_{\text{mag}} \propto \langle \mu \perp \vec{P} \times \vec{Q} \rangle^2.
$$

**Figure 2** | Neutron polarization analysis data measured at 3 K using D7 (ILL). (a) The non-spin-flip, i.e., NSF, channel. This channel measures nuclear scattering and magnetic scattering from moment components parallel to $\vec{P}$. (b) The spin-flip, i.e., SF, channel. This channel measures magnetic scattering from moment components lying in the scattering plane. Here the same colour code is used for both intensities. In this study, the neutron polarization, $\vec{P}$, is perpendicular to the scattering plane ($H, 0, L$).

**Figure 3** | Temperature dependence and reciprocal space maps in zero field using IN12 (ILL). (a) Temperature dependent $L$-scans at ($H_{\text{max}}, 0, L$). To correctly monitor the $L$-scans we first located the $H_{\text{max}}$ for $I_C$ at each temperature. (b) L-scans at representative temperatures. The intensity is vertically shifted for clarity. The solid lines are Lorentzian fits. Error bars are statistical errors. To clearly display the existence of the magnetic diffuse scattering as shown in (a) and (b), the observed magnetic intensity above 3.8 K is multiplied by 20. (c-e) $Q$-map in the ($H, 0, L$) scattering plane at 1.7, 3 and 7 K, respectively. The ellipse in (e) represents the expected $Q$-resolution.
In our study, the \(a\) and \(b\) axes are equivalent in the tetragonal symmetry. There is no magnetic signal in the NSF channel (Fig. 2a), therefore, the moments lie in the \((H, L)\) plane, mainly along the crystallographic \(c\) axis\(^5\) consistent with the powder neutron-diffraction study\(^5\). The uniaxial moment direction determined for the two spin states may be attributed to the Ising property of Er ions with a dominant crystal-field ground state \(|\pm 15/2\rangle\) which leads to a high magnetic anisotropy along the \(c\) axis\(^5\). The observed propagation vectors differ from those proposed in the neutron-powder-diffraction studies\(^5,51\) where positive and negative momenta cannot technically be differentiated.

**Reciprocal space maps and temperature-dependent phase diagram.** Figure 3 shows the temperature variation of the magnetic Bragg peaks. Only the \(I_C\) peaks are visible at the lowest temperature 1.7 K (Fig. 3c) in this study. As shown in Figs. 3a and b, when the temperature is increased there is a slight decrease in the peak intensities and the fractional \(L\). The intensities drop sharply at \(2.9\) K, and the \(I_C\) peak suddenly appears (Fig. 3d). The intensities for \(I_C\) continue to decrease gradually with increasing temperature while \(I_C\) intensity rises sharply to reach a maximum at \(3.2\) K before rapidly decreasing. The peaks cease to be sharp resolvable features and combine to become a weak diffuse peak above \(T_N \sim 4.2\) K (Fig. 3e). The temperature-dependent intensities are summarized in Fig. 4, showing a phase diagram that may be divided into three clear regimes.

**Magnetic-field dependent phase diagram.** The magnetic-field dependence of the intensities were measured at 3 K, which is the point where the intensities of \(I_C\) and \(I_C\) are roughly equivalent as

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**Figure 4 | Temperature-dependent phase diagram of the two spin states in zero field.** Integrated magnetic intensity from the Q-scans versus temperature. The phase diagram clearly shows three regimes. We attribute regime I to the AFM state from purely localized 4f moments; regime II to a mixture of localized 4f moments with a SDW from weakly-pinned collective spins in the valence bands (details in the text); and regime III to large amplitude short-ranged spin orders. The error bars are from Lorentzian fits.

**Figure 5 | Magnetic-field dependent phase diagram of the two spin states at 3 K.** (a) \(H\)-scans across the \(I_C\) peak with increasing magnetic field. (b) \(H_{\text{max}}\) (obtained from our Lorentzian fits) of the \(H\)-scans around \(I_C\) (diamonds) and nuclear \((2, 0, 0)\) (triangles) Bragg peaks as a function of both increasing (solid symbols) and decreasing (void symbols) magnetic field (directions as marked). (c) \(L\)-scans at \((H_{\text{max}}, 0, L)\) with increasing magnetic field, with \(H_{\text{max}}\) taken from (b). (d) Integrated magnetic intensities of the \(I_C\) (circles) and \(I_C\) (squares) peaks (along the \(L\)-direction) and of the \(I_C\) (diamonds) peaks (along the \(H\)-direction) as a function of increasing (solid symbols) and decreasing (void symbols) magnetic field. The lines are guides to the eye. The error bars are from Lorentzian fits.
shown in Fig. 4. The results are summarized in Fig. 5. The I_C intensities remain stable with increasing field until a threshold value of 0.1 T. Then they rapidly decrease as the field is increased up to 0.17 T (0.01 meV) (Figs. 5a, c, d), accompanied by an obvious shift of the I_C-Q-position from 1.436(1) to 1.442(1) (Figs. 5b and 6a). The L position keeps constant (Fig. 5c). In contrast, there are no corresponding changes in the intensities or positions of I_6 (Figs. 5c and 6b) or the underlying lattice, represented by the (2, 0, 0) nuclear Bragg reflection as shown in Fig. 5b. This indicates that the I_6 peaks are connected relatively closer with the underlying lattice, while the I_C peaks appear to be relatively independent. This sharp contrast implies that the electronic origins of the two sets of peaks are completely different in nature.

**Discussion**

Our data show evidence for different electronic origins of the magnetic peaks. We observe that the I_6 peaks persist down to the lowest temperature (~1.7 K) in this study, and they do not change position with field (Fig. 5c), and their intensities are constant below the threshold field (Fig. 5d). This is consistent with a ground-state AFM structure due to localized moments from the 4f electrons that are tightly bound within the Er atoms.

We believe that the I_C peaks are due to a SDW state from the following observations: (1) The I_C peaks are suppressed with a relatively weak magnetic field of 0.1 T (~0.006 meV), showing a fragile electronic instability that closely resembles the behaviour of CDWs driven by the electron-phonon interactions resulting from a lattice distortion\(^{24,44}\). It is stressed that the tetragonal symmetry of the crystal lattice is reserved in the whole studied temperature range, i.e., there is no any appreciable structural phase transition. In addition, our neutron polarization analysis confirms that no such CDWs exist in ErPd\(_2\)Si\(_2\) due to the absence of NSF scattering at the incommensurate position; (2) The peak positions shift with a change of the applied field, independently of the underlying lattice (Fig. 5b), behaving more like a fragile Wigner-electron-crystal\(^{58}\). This indicates that the electrons responsible for the magnetic order are able to move easily in the crystal like those freely-distributed for a metallic bonding; (3) It is unlikely that localized 4f electrons in ErPd\(_2\)Si\(_2\) would form two competing long-range magnetic states with the same moment direction within one pure tetragonal phase in the absence of magnetic contributions from the Pd and Si ions; (4) The resistivity of single-crystal ErPd\(_2\)Si\(_2\) (Fig. 7) displays no appreciable response to the appearance of I_C peaks. (5) The I_C intensity is not restored on removing the magnetic field, but indeed is recovered if the temperature is raised to 10 K (above \(T_N\)) for ~3 mins and then cooled to
3 K. By contrast, the \( I_2 \) intensity not only remains on the application of a field, it increases after removing the field. This clearly indicates that \( I_2 \) and \( I_C \) have different electron origins. Combining these five points leads us to propose that the \( I_C \) peaks are due to a SDW state, which is a consequence of the weak (in view of the small threshold field) electron-electron interactions\(^{33}\) mediated by the 4f moments\(^{40}\), plausibly associated with the itinerant conduction electrons near the Fermi level.

Figures 3 and 4 show clearly the coexistence and competition between the two modulated orders, which can be explained through a mechanism where conduction electrons contribute to both a SDW state, through Fermi surface nestings and s-f couplings\(^{49}\), and to mediating the dominant RKKY interactions between localized 4f moments\(^{40}\). These two functions are in general competitive, leading to a breakdown of the SDW state as the local-moment magnetism starts to fully develop below ~2.9 K. A similar breakdown also occurs in the field dependence, where the fragile SDW state is suppressed above ~0.17 T. Once above the field threshold, those conduction electrons that contribute to the SDW state then decouple and participate in the RKKY exchanges. The net result is that the \( I_C \) positions change, and their intensities are suppressed and unable to regain the original values when the field is removed (Figs. 5b, 5d, 6c and 6d), whereas the \( I_2 \) intensities grow with increasing field above ~0.17 T. Most importantly, the \( I_2 \) intensities almost linearly increase while decreasing the strength of magnetic field. The sharp suppression of the SDW state (Figs. 5a, 6c and 6d) at the threshold magnetic field is ascribed to the weak s-f couplings. The resistivity (Fig. 7) is insensitive to the change of the two spin states because the 4f electrons responsible for the dominant magnetic state are localized and seldom participate in the electrical conductivity. The collectively-organized conduction electrons responsible for the weakly-pinned low-energy SDW state tend to be delocalized due to the extremely small threshold magnetic field and begin to carry current in a small potential difference.

To summarize, we have shown that two incommensurate magnetic states, with different modulations propagating at \( Q_+ = (H \pm 0.557(1), 0, L \pm 0.150(1)) \) and \( Q_C = (H \pm 0.564(1), 0, L) \), respectively, but possessing the same moment direction, exist in an ErPd\(_2\)Si\(_2\) single crystal. We have established their temperature and magnetic-field dependent phase diagrams. We show that both states not only co-exist at ~3.2 – 4.2 K, but they are also in competition at ~2.9 – 3.2 K. One magnetic state correlates with the underlying lattice insofar as both are independent of the applied magnetic field in this study. In contrast, the other magnetic state is so delicate that a modest magnetic field of ~0.17 T can suppress it, and it is not recovered on releasing the field. We thus propose that the two states have different electronic origins. One state corresponds to the localized unpaired 4f electrons associated with the Er atoms, mediated mostly by the RKKY interactions, whereas the second one is a SDW state stemming from the conduction electrons, derived probably by the Fermi-surface nesting and/or their coupling to the local moments. We thus propose that ErPd\(_2\)Si\(_2\) represents a prototypical model system for simultaneously studying the interesting behaviors of itinerant and localized moments well-separated in one compound.

Methods

Resistivity measurements and in-house X-ray powder diffraction. The growth and in-house characterizations of ErPd\(_2\)Si\(_2\) single crystals were previously reported\(^{54,55}\). The electrical resistivity of a bar-shaped single crystal by standard dc four-probe technique was measured on a commercial physical property measurement system (PPMS), and a powder X-ray diffraction study was performed on an in-house diffractometer with a 20 step size of 0.005° at 300 K, employing the copper \( K_\alpha \) = 1.5406(9) Å radiation. The powder diffraction data were analyzed by the Fullprof Suite\(^{56}\).

Crystal quality and alignment. We selected a single piece with a mass of ~1.12 g for the neutron scattering studies, which was oriented in the (H, 0, L) scattering plane of the tetragonal symmetry with the neutron Laue diffractometer, OrientExpress\(^{46}\), at the Institut Laue-Langevin (ILL), Grenoble, France. The mosaic of this single crystal is 0.48(1)° full width at half maximum for the nuclear (2, 0, 0) Bragg reflection at ~230 K. Throughout this paper, the wave vector \( Q_\text{det}(\alpha) = (\frac{a}{2 \pi} Q_H, \frac{b}{2 \pi} Q_K, \frac{c}{2 \pi} Q_L) \) is defined in units of r.l.u., where \( a, b, \) and \( c \) are the relevant lattice parameters referring to the tetragonal unit cell.

Neutron polarization analysis. Uniaxial longitudinal neutron polarization analysis was performed at the D7 (ILL) diffractometer with incident vertically-polarized neutron spins with \( \lambda = 4.8 \) Å. In this polarization setup, the NSF channel collects neutron scattering intensities from the nuclear Bragg reflections and the out-of-plane magnetic moments along the <001> direction, while the SF channel records the magnetic intensity from the in-plane moments that are perpendicular to both the b axis and the scattering vector \( Q^\parallel \).

Unpolarized neutron scattering. Elastic neutron scattering measurements were carried out on the same oriented sample at the IN12 (ILL) cold triple-axis spectrometer with a vertical moderate magnetic field (up to 3.5 T) parallel to the [010] (or [100]) direction and fixed final energy at 10.03 meV. The single crystal was top-loaded, and the beam collimation throughout the experiment was kept at open-30°-sample-40°-open.

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