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Metamagnetic Behavior and Kondo Breakdown in Heavy-Fermion CeFePO

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We report that nonmagnetic heavy-fermion (HF) iron oxypnictide CeFePO with two-dimensional XY-type anisotropy shows a metamagnetic behavior at the metamagnetic field $H_M \approx 4$ T perpendicular to the $c$ axis and that a critical behavior is observed around $H_M$. Although the magnetic character is entirely different from that in other Ce-based HF metamagnets, $H_M$ in these metamagnets is linearly proportional to the inverse of the effective mass, or to the temperature where the susceptibility shows a peak. This finding suggests that $H_M$ is a magnetic field breaking the local Kondo singlet, and the critical behavior around $H_M$ is driven by the Kondo breakdown accompanied by the Fermi-surface instability.

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Metamagnetism is represented by a sudden increase in magnetization with increasing an applied field. In heavy-fermion (HF) systems, CeRu$_2$Si$_2$ with the tetragonal ThCr$_2$Si$_2$ structure shows the metamagnetic behavior at about 7.7 T when a magnetic field ($H$) is applied parallel to the $c$ axis. Although various experiments as well as theoretical studies have been carried out [1,2], the mechanism is still controversial. In order to understand the metamagnetic behavior in HF systems, it might be desired to investigate new metamagnetic compounds.

The iron oxypnictide CeFePO is a related material of the iron-based superconductor LaFePO [3,4]. They possess the same two-dimensional layered structure, stacking Ce(La)O and FeP layers alternately. Brüning et al. reported that CeFePO is a magnetically nonordered HF metal with a Sommerfeld coefficient $\gamma = 700$ mJ/(mol K$^2$) [5]. At present, it is difficult to synthesize large single crystals of CeFePO for NMR measurements, but $^{31}$P-NMR can probe in-plane and out-of-plane magnetic response separately using $c$-axis aligned polycrystalline samples. Here we report novel metamagnetic behavior observed in $H$ $\perp c$, and suggest that metamagnetism of Ce-based HF compounds is driven by Kondo breakdown (drastic reduction of $c$-$f$ hybridization) as clarified experimentally.

The polycrystalline CeFePO was synthesized by solid-state reaction [4]. Basic properties are consistent with the previous report [5]. To measure anisotropic magnetic properties of CeFePO, the samples were uniaxially aligned using a magnetic field [6]. The polycrystalline CeFePO was ground into powder, mixed with stycast 1266, and was rotated in the external field of 1.4 T while the stycast cures. The $c$ axis of the sample is nicely aligned, which is shown from the angle dependence of $^{31}$P-NMR spectra (see in the inset of Fig. 1), and $^{31}$P-NMR measurement was performed on the sample.

Figure 1 shows $H$-swept NMR spectra in $H \parallel c$ and $H \perp c$ obtained at 31.4 MHz and various temperatures ($T$). The resonance peak for $H \parallel c$ is almost $T$ independent, but the peak for $H \perp c$ shows the characteristic $T$ dependence originating from $\chi(T)$. The Knight shift $K_L$ was determined from the peak field of the $^{31}$P NMR spectrum obtained in $H$ perpendicular (parallel) to the $c$ axis. $K = 0$ was determined by reference material H$_3$PO$_4$. $K_L(T, H) (i = \perp$ and $\parallel)$, which is the measure of the local susceptibility at the nuclear site, is defined as

$$K_L(T, H) = \frac{K_0 - K_{res}}{K_{res}} \approx \frac{M_L(T, H_{res})}{M_{res}},$$

(1)

![FIG. 1 (color online). (Main panel) $T$ dependence of $H$-swept NMR spectra at 31.7 MHz for $H \perp c$ (solid line) and $H \parallel c$ (broken line). $K = 0$ was determined by reference material H$_3$PO$_4$. (Inset) Angle dependence of $H$-swept NMR spectra at 31.7 MHz measured at 20 K. $\theta$ is the angle between magnetic field and $c$ axis. Solid line is corresponding to fitting line.](image-url)
where \( H_{\text{res}} \) are magnetic fields at resonance peaks, \( H_0 \) and \( \omega_0 \) are the resonance field and frequency of bare \(^{31}\text{P} \) nucleus and have the relation of \( \omega_0 = \gamma_n H_0 \) with gyromagnetic ratio \( \gamma_n \), and \( M_c(T, H_{\text{res}}) \) is the magnetization under \( H_{\text{res}} \) (i = \perp \) and \( \parallel \) ) at \( T \). \( K_{\parallel} \) is almost independent of \( T \) and \( H \), whereas \( K_{\perp} \) shows strong \( T \) dependence originating from the Curie-Weiss behavior of \( \chi(T) \) above 10 K as shown in Fig. 2(a). The anisotropic Knight shift suggests that static spin properties possess \( XY \)-type spin anisotropy. It should be noted that \( K_{\perp} \) exhibits \( H \) dependence below 4 K and above 2 T, indicative of a nonlinear relation between \( M_{\parallel} \) and \( H \). Using the hyperfine coupling constant \( ^{31}A_{\text{hf}} = 0.2 \, T/\mu_B \) which is estimated from the plot between isotropic component of \( K \) and \( \chi(T) \) above 10 K (not shown), we can plot \( M_c(H) \) against \( H \) in Fig. 2(b). \( M_{\parallel}(H) \) becomes superlinear against \( H \) at 0.1 K, which is the hallmark of metamagnetism, whereas \( M_{\perp}(H) \) is linear up to 6.2 T, which is again highly anisotropic.

Next, we focus on \( T \) and \( H \) dependence of low-energy spin dynamics probed with the nuclear spin-lattice relaxation rate \( 1/T_1 \). \( 1/T_1 \) of \(^{31}\text{P} \) was measured at each resonance peak by the saturation-recovery method, and was uniquely determined by a single component in whole measured range. The inset of Fig. 3 shows \( T \) dependence of \( 1/T_1 \) at low field \( \mu_0 H \approx 0.6 \, T \) parallel and perpendicular to the c axis. Below 1.5 K, \( 1/T_1 \) as well as \( K \) along both directions becomes constant, indicative of the formation of a Fermi-liquid (FL) state of heavy electrons. In general, \( 1/T_1 \) probes spin fluctuations perpendicular to applied \( H \), and thus \( 1/T_1 \) in \( H \parallel c \) and \( H \perp c \) are described as

\[
(1/T_1)_{H\parallel c} = 2(\mu_0 \gamma_n)^2 \sum_q |H_{\perp}(q, \omega_{\text{res}})|^2
\]

\[
\propto 2A^2 \sum_q |S_{\perp}(q, \omega \approx 0)|^2, \text{ and}
\]

\[
(1/T_1)_{H\perp c} = (\mu_0 \gamma_n)^2 \sum_q [H_{\parallel}(q, \omega_{\text{res}})]^2 + |H_{\perp}(q, \omega_{\text{res}})|^2
\]

\[
\propto A^2 \sum_q |S_{\parallel}(q, \omega \approx 0)|^2 + |S_{\perp}(q, \omega \approx 0)|^2.
\]

Here \( |X(\omega)| \) denotes the power spectral density of a time-dependent random variable \( X(t) \), and \( A \) is assumed to be \( q \) independent due to the metallic state. From these equations, we can decompose spin fluctuations along each direction as shown in the main panel of Fig. 3. \( \sum_q S_{\perp}(q, \omega \approx 0)^2 \) is dominant at low \( T \), since \( (1/T_1 T)_{H\parallel c} \) is almost twice larger than \( (1/T_1 T)_{H\perp c} \). This indicates that the spin dynamics also possess \( XY \)-type anisotropy. The \( XY \)-type spin fluctuations have the predominance of ferromagnetic (FM) correlations as inferred from the Korringa relation between \( (1/T_1 T)_{H\parallel c} \) and \( K_{\perp} \) in low-T FL state, which is consistent with the previous \(^{31}\text{P}-\text{NMR} \) result \cite{5} and with the experimental facts that CeFePO is close to FM instability \cite{7,8}.

The evolution of the spin dynamics against \( H \) was investigated for both directions. Figure 4 shows \( T \) dependence of \( (1/T_1 T)_{H\perp c} \) below 4 T (approximately the metamagnetic field, \( H_{\text{MM}} \) (a) and above 4 T (b). Although \( (1/T_1 T)_{H\parallel c} \) does not depend on \( H \) up to 6.2 T as shown in the inset of Fig. 3, \( (1/T_1 T)_{H\parallel c} \) changes significantly by \( H \) as shown in Figs. 4(a) and 4(b). \( H \) dependence of

\[
\Sigma \sum_{q}(\omega_{\text{res}})^2
\]

\[
\Sigma \sum_{q}(\omega_{\text{res}})^2
\]

\[
\Sigma \sum_{q}(\omega_{\text{res}})^2
\]

\[
\Sigma \sum_{q}(\omega_{\text{res}})^2
\]
(1/T_1T)_{H \perp c} at 0.1 and 1.5 K is shown in Fig. 4(c). (1/T_1T)_{H \perp c} shows a distinct maximum at H_M, suggesting that the enhancement of the density of states (DOS) is related to metamagnetic behavior. If we assume that 1/T_1T \propto N(E_F)^2, N(E_F) at H_M is almost 1.5 times larger than N(E_F) at 0 T. However, it is noteworthy that non-FL behavior characterized by continuous increase in (1/T_1T)_{H \perp c} with decreasing T [broken lines are shown in (b)] was observed in a narrow field region intervening between low-field paramagnetic (PM) state and high-field polarized PM state above 6 T.

To investigate such evolution of FSs by H, we performed the ab initio band-structure calculation in the paramagnetic state of CeFePO by using the WIEN2K package [10]. FSs in the low-field region are composed of itinerant Ce 4f electrons as shown in Fig. 5(a). The large FS shows the characteristic neck structures around X-R at the Brillouin zone boundary, at which boundary electrons have small Fermi velocity or heavy electron mass. The orbital character is dominated by the j_z = \pm 1/2 component in the j = 5/2 multiplet of 4f orbitals, as seen in Figs. 5(a) and 5(c) [11]. These features of Fermi surface imply that the low-field HF state possesses the small q magnetic correlations and their in-plane component is much larger than the out-of-plane component, in good agreement with the experimental results. The band calculation also shows that applied field pinches off the neck FSs around R and X in order, that is, “the field-induced Lifshitz transition” appears. This is accompanied by a drastic change in DOS at the Fermi level, which can be a driving force for the
metamagnetic transition and non-FL behavior around $H_M$ [12,13]. With applying higher $H$, 4f electrons are localized, and thus the FSs become small. The resultant FSs are shown in Fig. 5(b), familiar in the iron-based superconductors [14]. Thus the scenario of the field-induced Lifshitz transition can link with the nature in the metamagnetic transition in CeFePO.

Here, we compare the present results with CeRu$_2$Si$_2$, one of the most well-known metamagnetic compounds. Although both compounds show similar $T$ and $H$ dependence of Knight shift and $1/T_1T$ in $H$ parallel to the magnetic easy axis [15], as well as the similar $H$-$T$ phase diagram defined by $T_{\text{max}}$ [16] as shown in Fig. 4(d), magnetic properties are quite different. For example, magnetic easy axis is different between CeFePO and CeRu$_2$Si$_2$: CeFePO possesses two-dimensional XY-type spin anisotropy, whereas CeRu$_2$Si$_2$ possesses Ising-type spin anisotropy [17]. As a result, the ground states of the crystal-field level are different and their metamagnetic behavior is observed in different directions. In addition, dominant magnetic fluctuations in CeFePO differ from those in CeRu$_2$Si$_2$. It is reported that CeRu$_2$Si$_2$ is located close to antiferromagnetic instability accompanied with FM fluctuations [18,19].

Figure 6 shows the relationship between the metamagnetic field $H_M$ and the temperature where the bulk susceptibility shows a maximum $T_{\text{max}}$ or inverse of Sommerfeld coefficient $\gamma$ at $H = 0$ for CeCu$_6$, CeFePO, and CeRu$_2$Si$_2$ with doped and pressurized systems [5,17,20–23]. It deserves mention that the linear relation holds between the two quantities, notwithstanding that the three compounds possess totally different crystal structures and magnetic properties. Since $T_{\text{max}}$ is regarded as a Kondo temperature $T_K$ and roughly speaking, the relation of $\gamma T_K = \text{const}$ holds in the HF state, these facts indicate that $H_M$ is merely related to the local Kondo singlet energy $T_{\text{max}}$ and is not linked with the magnetic fluctuations originating from the intersite coupling between neighboring Ce ions and/or the nesting between the “large” FS. The experimental fact that $H_M$ is linearly proportional to $T_K$ in Fig. 6 strongly suggests that the metamagnetic behavior is linked with the Kondo breakdown [24]. Therefore, in the Ce-based magnets, the Kondo breakdown and the Fermi-surface instability accompanied by the drastic change of DOS occur almost simultaneously around $H_M$, which can induce novel non-FL behavior.

In summary, we performed $^{31}$P-NMR in the uniaxially aligned CeFePO and found that CeFePO possesses two-dimensional XY-type FM fluctuations, and shows metamagnetic behavior when $H$ is applied to $H \perp c$ below 5 K, accompanied with non-FL behavior around metamagnetic field $H_M \approx 4$ T. As far as we know, this is a first example that the metamagnetic behavior occurs in a nonmagnetic Ce-based HF compound with the $XY$-type spin anisotropy. From the band calculation and the comparison with other Ce-based metamagnets, we
claim that $H_M$ is a magnetic field breaking the local Kondo singlet, which is determined with the intrasite coupling between Ce-4$f$ and conduction electrons, and that the FSs change drastically due to the Kondo breakdown.

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[11] To obtain the $J$-resolved Fermi surface, we applied the WANNIER90 code [25] via the WIEN2WANNIER interface [26], on the basis of the Ce 4$f$ and Fe 3$d$ orbitals. Figure 5(b) was calculated from the obtained tight-binding Hamiltonian under the condition that one $f$ electron per Ce atom is localized.