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Kyoto University
Metamagnetic Behavior and Kondo Breakdown in Heavy-Fermion CeFePO

S. Kitagawa,1,2,* H. Ikeda,1,2 Y. Nakai,1,2† T. Hattori,1 K. Ishida,1,2 Y. Kamihara,2,3 M. Hirano,4 and H. Hosono4,5
1Department of Physics, Graduate School of Science, Kyoto University, Kyoto 606-8502, Japan
2TRIP, JST, Sanbancho building, 5, Sanbancho, Chiyoda, Tokyo 102-0075, Japan
3Departments of Applied Physics and Physico-Informatics, Keio University, Kanagawa, 223-8522, Japan
4Frontier Research Center, Tokyo Institute of Technology, Yokohama, 226-8503, Japan
5Materials and Structures Laboratory, Tokyo Institute of Technology, Yokohama, 226-8503, Japan

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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, CeRu2Si2 with the tetragonal ThCr2Si2 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/\((\text{mol K}^2) \) [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 \( K_{\perp(\parallel)} \) 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 \( \text{H}_3\text{PO}_4 \). \( K_i(T, H) \) \((i = \perp \text{ and } \parallel)\), which is the measure of the local susceptibility at the nuclear site, is defined as

\[
K_i(T, H_{\text{res}}) = \left( \frac{H_0 - H_{\text{res}}}{H_{\text{res}}} \right) \frac{M_i(T, H_{\text{res}})}{H_{\text{res}}}, \tag{1}
\]

where \( M_i(T, H_{\text{res}}) \) is the moment of the \( i \)-type resonance, \( H_{\text{res}} \) is the magnetic field of the resonance peak, and \( H_0 \) is the magnetic field of the reference material (\( \text{H}_3\text{PO}_4 \)).
where $H_{\text{res}}$ are magnetic fields at resonance peaks, $H_0$ and $\omega_0$ are the resonance field and frequency of bare $^{31}$P nucleus and have the relation of $\omega_0 = \gamma_n H_0$ with gyromagnetic ratio $\gamma_n$, and $M_r(T, H_{\text{res}})$ 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 $K_{\parallel}$ exhibits $H$ dependence below 4 K and above 2 T, indicative of a nonlinear relation between $M_{\parallel}$ and $H$. The hyperfine coupling constant $A_{\text{hf}} = 0.2$ 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 $M_r(H)$ against $\chi$ in Fig. 2(b). $M_{\parallel}(H)$ becomes superlinear against $H$ at 0.1 K, which is the hallmark of metamagnetism, whereas $M_{\parallel}(H)$ 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 $^{31}$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 $1/T_1 H$ at low field $\mu_0 H \approx 0.6$ T parallel and perpendicular to the $c$ axis. Below 1.5 K, $1/T_1 H$ 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

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

\[
\mathcal{X} 2A^2 \sum_q |S(q, \omega \sim 0)|^2,
\]

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

\[
\mathcal{X} A^2 \sum_q \left[ |S(q, \omega \sim 0)|^2 + |S(q, \omega \sim 0)|^2 \right].
\]

(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. $\sum_q |S(q, \omega \sim 0)|^2$ is dominant at low $T$, since $(1/T_1 H_{\parallel c})$ is almost twice larger than $(1/T_1 H_{\parallel c})$ at low-$T$ FL state, which is consistent with the previous $^{31}$P-NMR result [5] and with the experimental facts that CeFePO is close to FM instability [7,8].

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

FIG. 2 (color online). (a) $T$ dependence of the Knight shift determined at the peaks of $H \perp c$ and $H \parallel c$ spectra obtained at various $H$. Strong anisotropy of Knight shift suggests that static spin properties possess $XY$-type spin anisotropy. (b) $H$ dependence of magnetization $M_r(H)$ ($i = \perp$ and $\parallel$) using the relation of $M_r(H) = K_i(H) H_{\text{res}}/A_{\text{hf}}$. Solid and broken lines are guide to eyes. $M_{\perp}(H)$ suddenly increases with increasing $H$ and deviates from linear relation in the field range of 3–5 T, which is a definition of a metamagnetic behavior, while such a behavior was not observed in $M_{\parallel}(H)$ up to 6.2 T and down to 1.5 K.

FIG. 3 (color online). (Main panel) $T$ dependence of low-energy spin fluctuations parallel and perpendicular to the $c$ axis at $\approx 0.6$ T evaluated with $1/T_1 H$ measured in $H \perp c$ and $H \parallel c$ [see Eq. (2)]. The in-plane spin fluctuations are dominant at low $T$, suggesting that the spin dynamics also possess $XY$-type anisotropy. (Inset) $T$ dependence of $(1/T_1 H)$ at 10.3 MHz ($\approx 0.6$ T) for $H \perp c$ and at 10.3 MHz ($\approx 0.6$ T), 31.7 MHz ($\approx 1.8$ T), and 107.2 MHz ($\approx 6.2$ T) for $H \parallel c$. $(1/T_1 H_{\parallel c})$ is independent of $H$ up to 6.2 T.

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\( (1/T_1)_H \) at 0.1 and 1.5 K is shown in Fig. 4(c). \( (1/T_1)_H \) shows a distinct maximum at around \( H_M \), suggesting that the enhancement of the density of states (DOS) is related to metamagnetic behavior. If we assume that \( 1/T_1 \propto N(E_F)^2 \), \( N(E_F) \) 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 a continuous increase of \( (1/T_1)_H \) 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 \textit{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 \( j_z = 1/2 \) 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 $H$, 4$f$ 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/$T_1 T$ 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 metamagnets, 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-$4f$ and conduction electrons, and that the FSs change drastically due to the Kondo breakdown.

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*shunsaku@scphys.kyoto-u.ac.jp
†Present address: Graduate School of Science, Tokyo Metropolitan University, Hachioji, Tokyo 192-0397, Japan.

[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 $4f$ and Fe $3d$ orbitals. Figure 5(b) was calculated from the obtained tight-binding Hamiltonian under the condition that one $f$ electron per Ce atom is localized.