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 $K_{\perp(\parallel)}$ 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 $H_3PO_4$. $K_i(T, H)$ ($i = \perp$ and $\parallel$), which is the measure of the local susceptibility at the nuclear site, is defined as

$$K_i(T, H_{\text{res}}) = \left( \frac{H_0 - H_{\text{res}}}{H_{\text{res}}} \right)_{\omega = \omega_0} \approx \frac{M_i(T, H_{\text{res}})}{H_{\text{res}}}, \quad (1)$$

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 $H_3PO_4$. (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.
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 $\omega_0 = \gamma_0 H_0$ with gyromagnetic ratio $\gamma_0$, and $M_s(T, H_{\text{res}})$ is the magnetization under $H_{\text{res}}$ ($i = \perp$ and $\parallel$) at $T$. $K_{\perp}$ is almost independent of $T$ and $H$, whereas $K_{\parallel}$ 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 $M_{\perp}$ and $H$. Using the hyperfine coupling constant $A_{\text{hf}} = 2.0 T/\mu_B$, which is estimated from the plot between isotropic component of $K$ and $\chi(T)$ above 10 K (not shown), we can plot $M_s(H)$ against $H$ in Fig. 2(b). $M_{\perp}(H)$ 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, $1/T_1$ 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

\[
(1/T_1)_{H\parallel c} = 2(\mu_0 \gamma_n)^2 \sum_q |H_\perp(q, \omega_{\text{res}})|^2 \\
\propto 2A^2 \sum_q |S_\perp(q, \omega - 0)|^2, \quad \text{and} \\
(1/T_1)_{H\perp c} = (\mu_0 \gamma_n)^2 \sum_q [|H_\parallel(q, \omega_{\text{res}})|^2 + |H_\perp(q, \omega_{\text{res}})|^2] \\
\propto A^2 \sum_q [|S_\parallel(q, \omega - 0)|^2 + |S_\perp(q, \omega - 0)|^2].
\]

(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. $\sum_q |S_\perp(q, \omega - 0)|^2$ is dominant at low $T$, since $(1/T_1 H_{\text{res}})$ is almost twice larger than $(1/T_1 H_{\text{res}})$. 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 $(1/T_1)_{H\parallel c}$ and $K_{\perp}$ in 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 $(1/T_1 H)_{H\perp c}$ below 4 T (approximately the metamagnetic field, $H_M$) (a) and above 4 T (b). Although $(1/T_1 H_{\text{res}})$ does not depend on $H$ up to 6.2 T as shown in the inset of Fig. 3, $(1/T_1 H)_{H\perp c}$ 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 $M_s(H)$ ($i = \perp$ and $\parallel$) using the relation of $M_s(H) = K_s(H)H_{\text{res}}/A_{\text{hf}}$. 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 $1/T_1 H_{\text{res}}$ 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 $H \parallel c$. $(1/T_1 H_{\text{res}})$ is independent of $H$ up to 6.2 T.
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 in \( (1/T_1) \) with decreasing \( T \) [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. An enhanced \( (1/T_1) \) at 0.1 and 1.5 K is shown in Fig. 4(c). The enhanced 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 in \( (1/T_1) \) with decreasing \( T \) [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. An enhanced \( (1/T_1) \) at 0.1 and 1.5 K is shown in Fig. 4(c).

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. 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 \( \overline{X}-\overline{R} \) 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). 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 \( \overline{R} \) and \( \overline{X} \) 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
FIG. 5 (color online). Fermi surfaces (FSs) calculated with the ab initio band-structure calculation as Ce-4f electrons are itinerant (a), or localized (b), where the Fermi velocity is mapped on the FSs. (c) shows the FS colored by the $j_z$ character in the $j = 5/2$ multiplet of 4f orbitals, where $j$ is the total angular momentum. (d) the partial density of states. The Fermi level corresponds to 0 eV.

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 $1/T_1/T$ 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 $T_K$ 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 intransit 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.