Field-Induced Ordering in the Heavy Fermion Compound \( \text{YbCo}_2\text{Zn}_{20} \)

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Abstract. DC Magnetization of the heavy-fermion compound \( \text{YbCo}_2\text{Zn}_{20} \) was examined at very-low temperatures down to 0.07 K in the field up to 14.5 T. In addition to the nearly-isotropic metamagnetic behavior at 0.6 T, a new metamagnetic behavior is observed in the field variation of magnetization \( M(H) \) at 6 T only for \( H \parallel [111] \). For the same field direction, a clear kink appears in the temperature dependence of the magnetization \( M(T) \) for fields above 6 T. These results strongly suggest the existence of a new ordered phase induced by magnetic fields. The \( H-T \) phase diagram as well as the anisotropy in \( M(H) \) above \( \sim 2 \) T are best explained by a level crossing of the low-lying crystalline-electric-field states by magnetic fields.

1. Introduction

The majority of heavy fermion systems studied so far are Ce- or U-based compounds. Ytterbium with \( 4f^{13} \) electronic configuration, which is considered the hole counterpart of the \( 4f^1 \) configuration of Ce, also possesses the same potential to exhibit the heavy fermion behavior. While properties of Yb-based heavy fermions are in many respects similar to those of Ce-based systems owing to the electron-hole analogy, there appears to be some seeming differences between Yb and Ce based heavy fermions regarding the quantum critical behavior, as observed in \( \beta-\text{YbAlB}_4 \) [1] and \( \text{YbRh}_2\text{Si}_2 \) [2]. In this regards, it is of fundamental interest to study the Yb-based heavy fermion compounds which are in proximity to the quantum critical point.

Recently, a class of heavy fermion systems \( \text{YbT}_2\text{Zn}_{20}, \ (T = \text{Fe, Co, Ru, Rh, Os and Ir}) \), with a cubic CeCr\(_2\)Al\(_{20}\) type structure have been synthesized [3, 4]. All these compounds show a large electronic specific heat coefficient \( \gamma \) in excess of 500 mJ/mol-K\(^2\). The electrical resistivity \( \rho(T) \) also shows a strong \( T^2 \) dependence at low temperatures with the coefficient \( A \) exceeding \( 5\times10^{-2} \) \( \mu \Omega\text{cm/K}^2 \). Among the \( \text{YbT}_2\text{Zn}_{20} \) compounds, \( \text{YbCo}_2\text{Zn}_{20} \) is of particular interest because of its gigantic \( \gamma \) value as large as 7900 mJ/mol-K\(^2\). This compound exhibits a metamagnetic peak structure in the field dependence of the AC susceptibility \( \chi_{ac}(H) \) at a very low field of \( \mu_0H_m = 0.57 \) T applied along the [100] direction [5]. In particular, a magnetic phase has been reported to appear under a relatively low pressure of 1 GPa [6]. It is therefore considered that \( \text{YbCo}_2\text{Zn}_{20} \) is in close proximity to a quantum critical point. In this paper, we report on the low temperature magnetizations of a single crystal of \( \text{YbCo}_2\text{Zn}_{20} \) along the three principal field directions. We found that a distinct magnetic anisotropy develops in fields above 2 T with [111] being the...
Figure 1. Low field part of the magnetization $M(H)$ of YbCo$_2$Zn$_2$O$_{20}$ below 1 T along the three principle field directions [100] (circles), [110] (squares) and [111] (triangles) obtained at 0.07 K. The inset shows the field variation of the differential susceptibility $dM(H)/dH$.

Figure 2. $M(H)$ of YbCo$_2$Zn$_2$O$_{20}$ up to 14.5 T for $H \parallel [100]$ (circles), [110] (squares) and [111] (triangles) measured at 0.08 K. The inset displays $dM(H)/dH$ for $H \parallel [111]$ obtained at 0.08 K (solid triangles) and 0.4 K (open triangles).

2. Experimental Results and Discussion

Single-crystal samples of YbCo$_2$Zn$_2$O$_{20}$ were grown by the Zn-self-flux method. DC magnetization measurements down to 0.07 K in magnetic fields up to 14.5 T were performed using a capacitive Faraday magnetometer with a dilution refrigerator [8]. All the measurements were done in a gradient field of 5 T/m.

Figure 1 displays the field variation of the magnetization $M(H)$ of YbCo$_2$Zn$_2$O$_{20}$ below 1 T for $H \parallel [100]$ (solid circles), [110] (solid squares) and [111] (solid triangles) obtained at the base temperature of 0.07 K. A metamagnetic upturn is observed in $M(H)$ at around 0.6 T for all three field directions. This metamagnetic behavior can be seen more clearly as a peak structure in $dM(H)/dH$ as shown in the inset, which well reproduces the previous $\chi_{\text{ac}}(H)$ data in a [100] magnetic field. As can be seen from the figure, the peak position is nearly independent of the field directions implying that the anisotropy of this low field metamagnetism is very weak. This result is in contrast to the metamagnetic behavior seen in other non-magnetic heavy fermion systems. For instance, metamagnetism in CeRu$_2$Si$_2$ is driven only by the field component parallel to the $c$ axis [9]. Even in the cubic system YbIr$_2$Zn$_2$, the metamagnetic crossover field $H_m$ is anisotropic, with $\mu_0H_m = 9.7$ and 12 T for $H \parallel [100]$ and [110], respectively [10]. The nearly isotropic metamagnetism in YbCo$_2$Zn$_2$O$_{20}$ is probably due to the small critical field (0.6 T) in addition to the cubic symmetry.

Figure 2 shows $M(H)$ of YbCo$_2$Zn$_2$O$_{20}$ measured for the three field directions at $T = 0.08$ K. As can be seen from the figure, a distinct anisotropy develops above $\sim 2$ T: $M_{[100]} > M_{[110]} > M_{[111]}$. Remarkably, $M(H)$ for the hard axis [111] exhibits metamagnetic behavior at $\mu_0H_m = 6$ T, which is more clearly seen in the $dM(H)/dH$ plot as shown in the inset. The peak in $dM(H)/dH$,

hard axis. More importantly, we obtained evidence of a field-induced ordering along the [111] direction at low temperatures below 0.6 K [7].
which marks the metamagnetism, moves to the high field side with increasing $T$. The amplitude of the peak rapidly diminishes at $T = 0.4$ K where only a small anomaly remains around 7 T.

In order to further examine this high-field feature in the magnetization, we measured the temperature variation $M(T)$ in [111] magnetic fields and the results are shown in Fig. 3. As can be seen from the figure, we observed a clear kink (an upward bending) in $M(T)$ for fields above 6 T. The position of the kink in $M(T)$, defined as $T_0(H)$, moves to the higher temperature side with increasing $H$. In the inset of Fig. 4, we plotted both $H_m'(T)$ and $T_0(H)$ in a $H - T$ diagram. The results strongly suggest the presence of a field-induced ordered phase (FIOP) in fields parallel to [111]. It should be noticed that the positive slope of the phase boundary ($dT_0/dH > 0$) and the upward bending in $M(T)$ at $T_0$ are qualitatively in agreement with the Ehrenfest relation for a second-order phase transition [7].

The FIOP in Fig. 4 is reminiscent of a field-induced ordering in the Pr-based skutterudites PrOs$_4$Sb$_{12}$ and PrFe$_2$P$_{12}$ [11, 12, 13], in which a crossing among the Zeeman split low-lying crystalline-electric-field (CEF) states drives FIOP. We discuss this possibility in YbCo$_2$Zn$_{20}$, on the basis of a localized $f$ electron picture. The $J = 7/2$ multiplet of Yb$^{3+}$ splits into two doublets ($\Gamma_6$ and $\Gamma_7$) and a $\Gamma_8$ quartet in the cubic field. Magnetic moment values ($H \parallel [100]$) of these levers are 1.3, 1.7 and 2.1 $\mu_B$/Yb for $\Gamma_6$, $\Gamma_7$ and $\Gamma_8$ states, respectively. The observed magnetization in excess of 3 $\mu_B$/Yb thus implies that at least two CEF states are involved in the low-lying state. Actually, the specific heat divided by temperature, $C/T$, measured by Takeuchi et al. [14] revealed a shoulder structure around 3 K, which can be attributed to a Schottky peak due to a CEF excited state with the energy gap $\Delta/k_B$ below $\sim 10$ K. In order to reproduce the experimental results, we adopt the CEF scheme $\Gamma_6$ (0 K) - $\Gamma_8$ (6 K) - $\Gamma_7$ (27 K). The calculated field variation of the magnetization at 0.08 K is shown in Fig. 4, which well explains the observed magnetic anisotropy $M_{[100]} > M_{[110]} > M_{[111]}$. The CEF level scheme yields a crossing field of 10 T existing only for $H \parallel [111]$, in agreement with the FIOP whose transition temperature $T_0$ becomes highest at this field. Moreover, the broad peak in $M(T)$ appearing around 1.5 K...
in Fig. 3 can also be qualitatively understood by the CEF model [15]. From these results, we consider that the FIOP is due to a CEF level crossing in magnetic fields.

In general, Kondo spin fluctuations smear the CEF levels in heavy fermion systems. It is thus highly remarkable that a CEF level crossing drives the field-induced ordering in YbCo$_2$Zn$_{20}$. In this compound, we consider that Yb ions become close to a trivalent state under high magnetic fields. This would also be a consequence of the fact that YbCo$_2$Zn$_{20}$ is in proximity to QCP.

3. Summary
We measured DC magnetization of YbCo$_2$Zn$_{20}$ along the three principal field directions $H \parallel [100]$, [110] and [111]. In addition to the nearly isotropic metamagnetic behavior at 0.6 T, we obtained evidence of a field-induced ordered phase above 6 T for $H \parallel [111]$. The observed anisotropic phase diagram as well as the magnetic anisotropy strongly suggest that the field-induced ordered phase is driven by a crossing of low-lying CEF levels in a [111] magnetic field.

Acknowledgments
This work has been supported by a Grant-in-Aid for Scientific Research on Innovative Areas “Heavy Electrons” (No. 20102007) of the Ministry of Education, Culture, Sports and Technology, Japan and a Grant-in-Aid for Scientific Research (A) (No. 20244053) from the Japan Society for the Promotion of Science.

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