Possible critical behavior and field-induced magnetic transition in YbCo$_2$

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Abstract

The first study of electrical resistivity, magnetization and specific heat on YbCo$_2$ is reported. The measurements on single-phased sample of YbCo$_2$ bring no-evidence of magnetic ordering down to 0.3 K in zero magnetic field. The manifestations of low Kondo temperature are observed. It can explain evidently the very large electronic specific heat which places YbCo$_2$ among heavy fermion systems. Application of magnetic field revealed metamagnetic-like anomaly and field induced transition-like behavior. Despite the metamagnetic-like anomaly being weakly pronounced, the temperature-dependent measurements show obvious field induced-like transition under magnetic field above 2 T. Further, ferromagnetic character is exhibited as the transition temperature increases with increasing field. The origin of the observed properties is discussed in comparison and contrast with other RCo$_2$ and Yb-based heavy fermion compounds. It indicates that in the unique case of YbCo$_2$, the itinerant electron magnetism of Co 3$d$-electrons and the Kondo effect within the vicinity of quantum criticality of Yb 4$f$-local moments can both play a role.

Keywords: rare-earth element, magnetic transition, Yb-compounds, Kondo effect, heavy fermion
Introduction

Interest of Ce, Eu or Yb elements in intermetallic compounds are based on valence variety and \( c-f \) electron interactions which bring many interesting behaviors such as heavy fermion, non-BCS superconductivity, non-Fermi liquid, valence transition, quantum criticality etc... These phenomena are understood by considering the strength of \( c-f \) interaction which influences effectivity of RKKY interaction to promote magnetic ordering, whereas otherwise the Kondo effect overwhelms RKKY interaction. The \( c-f \) hybridization also results in fact that \( 4f \)-electrons of these elements can create local or itinerant magnetic moments and in many cases are in mixed valence state. These characteristics favor to form heavy fermion compounds with metamagnetic transition (MMT) as for example CeRu\(_2\)Si\(_2\), YbIr\(_2\)Zn\(_{20}\), YbRh\(_2\)Si\(_2\) and some rare earth borides\(^{1-4}\). These properties can be mapped by Doniach’s phase diagram. However, there are several compounds which possess properties hardly explained by above mentioned scenario. For example, fermi liquid state is expected to evolve well below Kondo screening but for CePd\(_3\) different correlation is observed\(^{5,6}\). It illustrates that not only by \( c-f \) hybridization can be used for description of these compounds but also \( d-f \) mixing need to be consider.

Compounds with potential of \( d-f \) mixing can be found among the \( RCo_2 \) compounds (\( R \) represents rare-earth element) which are known by 3\( d-4f \) magnetism. Compounds with Ce, Eu or Yb element can be involved into above mention problematic. In the literature, complex research study can be found only for CeCo\(_2\)\(^{7-11}\). The CeCo\(_2\) reflects different properties than other compounds in \( RCo_2 \) series, which are strong ferro/ferri-magnets with two magnetic sublattices created by itinerant Co and localized \( R \) magnetic moments\(^{12-16}\). For CeCo\(_2\), the Co 3\( d \) bands strongly hybridized with 4\( f \)-bands, which drives the compound almost to the nonmagnetic regime both for 4\( f \) and 3\( d \) electrons\(^{10}\). This brings our attention to the compounds with Eu and Yb in \( RCo_2 \) series which should be also affected by \( c-f \) and \( d-f \) interactions. In the literature can be found only two papers about YbCo\(_2\)\(^{17,18}\). They refer to the crystal structure and X-ray absorption measurement. No electrical resistivity, magnetization or specific heat data can be found for investigation of YbCo\(_2\)-magnetism. In this paper we bring results of characteristic measurements and analysis on successfully-prepared single phase YbCo\(_2\) polycrystalline samples. Long-range ordering at around 5 K which was previously theoretically predicted is not observed down to 0.3 K\(^{19}\). The signalized very low Kondo temperature in YbCo\(_2\) makes this compound unique not only among \( RCo_2 \) – series compounds but also in all the Yb-based compounds. Moreover, we observed field-induced magnetic transition in magnetization, electrical resistivity, and specific heat of YbCo\(_2\). It can be responsible for the observed large electronic specific heat contribution which witnesses the strong electron correlations.

Experimental

The YbCo\(_2\) was prepared from mixed powders of pure elements Co (3N) and Yb (3N). Pellets of mixed powders were sealed in Ta-tube and heated in an induction furnace under Ar flow up to 1200 °C for 5 minutes. Subsequently, the Ta-tube with the sample was annealed at 800 °C for 5 days. The brittle product of YbCo\(_2\) was consolidated by using Spark Plasma Sintering device (SPS, LABOX\textsuperscript{TM}110, Sinter Land). The sample was pressed up to 50 MPa and heated at 900 °C for 5 minutes under vacuum of \(~10^2\) Pa. The x-ray diffraction (performed on Miniflex, Rigaku equipped with a Cr-target) showed single phase of YbCo\(_2\) and confirmed
crystallization in the cubic MgCu2-type Laves phase with lattice parameter $a = 7.1135(2)$ Å (see Fig. 1). Stoichiometric composition was confirmed by scanning electron microscope (JSM-7001F, JEOL Ltd.) equipped by energy dispersive spectroscopy (EDS). The sample contained negligible amount of Yb$_2$O$_3$.

The magnetization measurement was performed using magnetic property measurement system with fields up to 7 T (MPMS 7T, Quantum Design Inc.). Standard electrical resistivity and specific heat measurement (with units J/mol.K per formula unit) were carried out by Physical Property Measurement System (PPMS, Quantum Design Inc.) under magnetic field up to 7 T. Low temperature measurement of specific heat down to 0.3 K was performed with using $^3$He insert for PPMS.

**Results and Analysis**

Temperature dependence of magnetic susceptibility measured at 1 T is depicted in Fig. 2. There is no magnetic transition down to 2 K, as was expected to occur at 5 K according to previous theoretical prediction in Ref. [19]. The results of Curie-Weiss fit in the paramagnetic region from 150 K to 300 K gives an effective magnetic moment of $\mu_{\text{eff}} = 6.16(5)$ $\mu_B$. This value is much larger than that expected for a free Yb$^{3+}$ ion of 4.56 $\mu_B$. It indicates that Co 3$d$-electrons also participate in the Curie-Weiss paramagnetism because the Yb$^{3+}$ valence state with magnetic moment was confirmed by X-ray absorption measurement on YbCo$_2$ sample prepared by high-pressure synthesis$^{18}$. Thus, we assume presence of Yb$^{3+}$ magnetic moment in value of 4.56 $\mu_B$ and calculate corresponding Co magnetic moment by formula $\mu_{\text{eff}} = \sqrt{\mu_{\text{Yb}}^2 + 2\mu_{\text{Co}}^2}$. The calculation yielded $\mu_{\text{Co}} = 2.94$ $\mu_B$. This value is similar with the value of the Co-effective magnetic moment observed for Y(Co$_{1-x}$Al$_x$)$_2$ in Ref [20]. The Curie-Weiss fit show negative paramagnetic Curie temperature $\theta_p = -71$ K pointing to an antiferromagnetic interaction in the paramagnetic region. On the other hand, negative $\theta_p$ does not necessarily indicate antiferromagnetic ordering for the case of itinerant electron magnetism. Indeed, negative $\theta_p$ is seen for nearly ferromagnetic Y(Co$_{1-x}$Al$_x$)$_2$ $^{20}$. Thus, the contribution of Co 3$d$ electrons may also be important in $\theta_p$ of YbCo$_2$ as well. The magnetization curves (see inset in Fig. 2) demonstrate paramagnetic behavior for temperatures above 20 K. On the other hand, magnetization curves measured at lower temperatures suggest nearly ferromagnetic-like behavior which can point to possible magnetic ordering at lower temperatures. The derivative of low-temperature magnetization curves shows a local maximum depicted in Fig. 3 which indicates the possibility of MMT. Among RCo$_2$ compounds, only YCo$_2$ and LuCo$_2$ show MMT out of paramagnetic state but the transition appears at high magnetic field ~ 70 T and is caused by Co sublattice$^{21, 22}$. It is quite contrasting with the present low field metamagnetic-like transition and no-step anomaly which we observed in the case of YbCo$_2$. Temperature dependence of magnetization possesses manifestation of magnetic kind of anomaly as is depicted in Fig. 4. The characteristic temperature of the anomaly is taken as the minimum of the derivative and application of magnetic field shifts the anomaly to higher temperatures. It is notable that a similar evolution of field-induced phase transition has been reported for YbCo$_2$Zn$_{20}$.$^{2,3,24}$ Its origin was interpreted by level-crossing of crystal-field states caused by application of magnetic field. Similarities with YbCo$_2$ are discussed in the following section.
The high temperature resistivity curve in YbCo₂ saturates as is depicted in Fig. 5. This saturation is caused by the spin fluctuation scattering and spin disorder contribution to electrical resistivity in the paramagnetic state as in other $RCO_2$ compounds. With decreasing temperature, the resistivity gradually decreases below 200 K and reaches a local minimum at around 15 K. The decrease corresponds to the reduction of 4f-orbital degeneracy due to the crystal field splitting. For further decreasing temperatures, the resistivity shows a small maximum at 8 K, below which the resistivity decreases rather rapidly. This rapid decrease can be attributed to the Kondo-lattice behavior, as is often observed in Yb compounds. Nevertheless, in comparison with other Yb-based compounds, the Kondo anomaly of YbCo₂ is situated at lower temperatures. This indicates very low Kondo temperature $T_K$. The local minimum from the Kondo anomaly is gradually suppressed by increasing of magnetic field and further, above 2 T, the change of curvature at low temperature gives suggestion of some kind of transition (see in Fig. 6 a). The derivative of electrical resistivity curves clearly shows a maximum, which indicates the existence of the magnetic-field induced-like transition (Fig. 6 b). The fit of the low temperature evolution is depicted in Fig. 7. The resistivity curve measured at 0 and 1 T have concave character and can’t be fitted by standard formula. Increasing of magnetic field induces convex character of resistivity curves. The fit can be provided for curves measured above 1 T. As is depicted in Fig. 7, the fit can be used for longer temperature range and the exponent of temperature also increases with increasing of magnetic field. The value of the fitted exponent starts at 1.2 and saturates around 1.6. It is less than 2 expected for Fermi liquid behavior and moreover the saturated value corresponds to theory of critical fluctuation of non-Fermi liquid for 3-dimensional ferromagnets which makes signature of critical behavior.

The specific heat measurement is depicted in Fig. 8. There is observed a peak around 2.3 K which originates from small amount of Yb₂O₃. The data below 1 K show increasing trend of specific heat, as shown in the inset of Fig. 8. Usually, at such low temperature nuclear contribution should be taken into account, but the observed anomaly has a different temperature evolution than $T^2$ expected for nuclear contribution and calculated nuclear heat capacity for each element in Ref [27] gives much smaller value. Therefore, the origin of increase in $C_p$ below 1 K is likely to be caused by other critical behavior. The specific heat also shows a broad bump with the maximum at 4.9 K. This anomaly corresponds well to the broad maximum in the electrical resistivity, thereby is probably caused by Kondo interaction. The magnetic contribution to the specific heat, $C_{mag}$, has been evaluated by subtracting the specific heat of a nonmagnetic reference YNi₂. We attempted to compare the $C_{mag}$ at low temperature with the Kondo model, calculated by Rajan. The result is shown in Fig. 9. The low temperature $C_{mag}$ has been accounted for by the 3fold or 2fold-degenerated Kondo model. This low degeneracy is attributed to the effect of crystal field splitting. Similar behavior can be found in Refs. [29, 30] for YbPdSb and YbRh₂Zn₂₀ compounds where the bump in low temperatures is interpreted as the Kondo screening ground state quartet of crystal electric field associated with very low Kondo temperature. On the other hand, the middle temperature range of $C_{mag}$ is well explained by the Schottky contribution from two quartets with the energy separation of $\Delta \sim 115$ K. This is different from the case of YbNi₂ where a doublet ground state, first excited quartet and second excited doublet has been presented. Fig. 10 shows temperature dependence of $C_{p}/T$ at low temperatures which has unexpectedly increasing character upon cooling and gives a large values of $C_{p}/T$. This behavior makes it difficult to evaluate the Sommerfeld coefficient, but indicates its large value. Increasing character of $C_{p}/T$ upon cooling (different than the nuclear
contribution since evolution doesn’t correspond to $C_\text{p} \sim T^2$) agrees with the absence of Fermi liquid dependence ($\rho \sim T^2$) in the electrical resistivity, as described above. These observations can be considered in relation to non-Fermi liquid behavior and close quantum criticality as were reported for example in YbRh$_2$Si$_2$, CeCu$_{6-x}$Ag$_x$ and YbCu$_{5-x}$Al$_x$ compounds$^{3,32,33}$.

It should be notice that the above-mentioned non-Fermi liquid systems are close to antiferromagnetic ordering, and application of magnetic fields can drive the system towards the nonmagnetic Fermi-liquid regime. In contrast, specific heat measurement of YbCo$_2$ under application of magnetic field up to 7 T revealed peak behavior as is depicted in Fig. 10. The peak is moving to higher temperatures with increasing magnetic field, which points to ferromagnetic character of this transition. This demonstrates the evolution of magnetic ordering induced by magnetic field, consistent with the anomaly in the electrical resistivity (shown in Fig. 6). Therefore, YbCo$_2$ can be regarded as very close to ferromagnetic quantum criticality. Here, it is possible to estimate the electronic specific heat coefficient ($\gamma$) for the data in magnetic field above 4 T by extrapolating $C_p/T$ data toward $T \to 0$ in Fig. 10. The $\gamma$ values are roughly obtained to be 1 to 2 J/mol.K$^2$. Electrical resistivity indicates gradual restoration of Fermi liquid behavior by magnetic field but it seems to be saturated at $n = 1.6$ pointing to ferromagnetic critical fluctuations. Nevertheless, it can be also caused by evaluation close to transition and Fermi liquid state can coexists with field-induced ferromagnetic state in lower temperatures.

Discussion

The susceptibility measurements and the results of Curie-Weiss fit gave higher value of $\mu_{\text{eff}}$ than expected for Yb$^{3+}$. It leads us to infer the existence of contribution of Co magnetic moment. This is a contrasting case compared to the mixed valence state of CeCo$_2$, where the magnetic character of both Ce and Co appears to be suppressed$^3$. The metamagnetic-like behavior in magnetization curves brings a question about similar origin of this behavior as has been observed in such heavy fermion metamagnetic compounds as CeRu$_2$Si$_2$, YbIr$_2$Zn$_{20}$, YbCu$_3$, UPd$_2$Al$_3$, URu$_2$Si$_2$, YbCo$_2$Zn$_{20}$, etc$^{1,2,33-36}$. The common parameter in these compounds is the interplay of Kondo and RKKY interaction which results in large electronic specific heat coefficient and change between local and itinerant magnetism by temperature or magnetic field. The localized magnetism in high temperatures changes to itinerant at low temperatures which is usually accompanied by a local maximum in susceptibility curve. Our measurements suggest the existence of Yb$^{3+}$ magnetic moment at high temperatures but no sign of local maximum in susceptibility curve down to 2 K is detected. Although it may be found at lower temperatures such as previously observed in YbCo$_2$Zn$_{20}$.$^2$ Another contrast between the observed anomaly in YbCo$_2$ and the above-mentioned compounds can be found in the temperature dependence. The MMT in heavy fermion compounds usually moves to lower temperatures or it shows almost temperature independent behavior. Our observed anomaly shifts to higher temperatures quickly and becomes broader.

From this point of view, our observed anomaly is not likely to be a heavy-fermion metamagnetism, but rather has ferromagnetic-like property. This indicate a polarized paramagnetic phase or ferromagnetic transition hidden at lower temperature than we could reach. In this case we should also mention that for $RCO_2$ ($R = $ Er, Ho, Dy), the first order magnetic phase transition has been observed due to the coupling of Co 3$d$-based itinerant electron magnetism and the $R$ 4$f$-based localized moment. The MMT is measured at a few
Kelvins above the Curie temperature with field hysteresis\(^{37}\). It is basically attributed to a field-induced itinerant-electron metamagnetism due to the Co-magnetic moment. Nevertheless, it is in contrast with the presented magnetization measurement on YbCo\(_2\) which doesn’t show hysteresis. In addition, the data of specific heat witness a huge electronic specific heat coefficient in YbCo\(_2\) roughly estimate in the range of 1 to 2 J/mol.K\(^2\) for 4-7 T. This indicates that heavy fermion behavior evolves below the observed anomaly. This result suggests that the magnetic transition most likely involves not only the 3\(d\) bands but the very heavy 4\(f\) quasiparticle band as well. It can be supported by results of fit of electrical resistivity curves where non-Fermi liquid behavior is observed through value of the exponent \(n\). The value is increasing by application of magnetic field but saturates at 1.6 which is close to 5/3 as predicted by theory for ferromagnetic correlation\(^{36}\). The critical state is also suggested by Wilson ratio which is between 1.4-1.6 for measurements at 3-5 T. The value is higher than 1 that corresponds to Fermi liquid state and less than 2 for Kondo state. Nevertheless, observed values are in agreement with values of others heavy fermion compounds shown in Ref\(^{38}\).

As is depicted in Fig. 10, the increasing character of \(Cp/T\) upon cooling down to 0.3 K doesn’t allow to definitely determine the gamma value at zero field but predicted large value which place YbCo\(_2\) between heavy fermion systems. This behavior is also observed in heavy-fermion systems such as YbRh\(_2\)Si\(_2\) due to its close vicinity of the quantum criticality\(^3\) or YbCo\(_2\)Zn\(_{20}\) where the very low Kondo temperature \(T_K = 1\) K needs to be considered together with crystal electric field (CEF)\(^2\). As it is mentioned above, the Kondo screening is presented in our sample and from manifestation in electrical resistivity and specific heat, the Kondo temperature is estimated to be as low as a few Kelvins. The low Kondo temperature can sometimes lead to the coexistence of magnetic ordering with the Kondo lattice formation like in the case of YbPdSb\(^{29}\).

Nevertheless, results from electrical resistivity, temperature dependence of magnetization and specific heat under application of magnetic field shows pronounced anomalies that suggest long-range ordering. The anomalies are similar with reported observations on YbCo\(_2\)Zn\(_{20}\)\(^{2,23,24}\). The field-induced ordered phase is present in measurement of magnetic field along the <111> crystal lattice. This type of long-range ordered phase is considered as field-induced antiferroquadrupole ordered phase and is understood within the CEF model with two degenerated lowest CEF levels\(^2\).

Up to now, we can’t definitely conclude about the origin of observed anomalies in YbCo\(_2\) which we summarized in the magnetic-like phase diagram in Fig. 11. The ferromagnetic-like behavior of the observed anomalies is very unusual in heavy fermion systems as most of compounds undergo antiferromagnetic ordering. It is likely that the heavy-fermion metamagnetism is coupled with the Co 3\(d\)-itinerant electron metamagnetism. The detail study at low temperatures as well as neutron diffraction experiments under fields is necessary to resolve origin of observed phenomena and investigation of grown single crystal would shed more light onto these issues as well.

Conclusions

The paper brings first complex study of YbCo\(_2\) by the electrical resistivity, specific heat and magnetization measurements. The Yb ion is indicated to be in the magnetic Yb\(^{3+}\) valence state together with Co magnetic moment in the order of 3 \(\mu_B/Co\). Low temperature measurements revealed that the Kondo effect contributed to electrical resistivity and specific
heat properties with evidence of low Kondo temperature. As has been discussed in comparison with other compounds, low Kondo temperature in heavy fermion materials with CEF can be responsible for critical properties and MMT.

The measurements under application of magnetic field revealed metamagnetic-like and ferromagnetic-like transitions. These manifested anomalies are similar with observations on YbCo$_2$Zn$_{20}$ which has MMT at very low temperatures and field-induced ordered phase. However, several missing connections make this comparison controversial such as the separation of MMT and field-induced ordered phase, or missing susceptibility maximum in YbCo$_2$. Another possible explanation can be that a ferromagnetic transition due to Co 3$d$ electrons is hidden at temperatures below 0.3 K which would be very sensitive to applied magnetic field. In any case, the very large electronic specific heat coefficient below the field-induced magnetic-like-transition temperatures suggests that the observed properties in YbCo$_2$ involve both: the itinerant electron magnetism of Co 3$d$-electrons and the Kondo effect within the vicinity of quantum criticality of Yb 4$f$-local moments. Partial support is brought by non-Fermi liquid behavior observed in electrical resistivity and from Wilson ratio.

The results indicate that YbCo$_2$ is a unique and attractive example for the case where both the 3$d$- and 4$f$-electrons play a role in the magnetic instability. Further experiments with single crystalline samples are desired.

Acknowledgments

This work was supported by the Grand-in-Aid from Japanese Society for the Promotion of Science, KAKENHI, No. P21322 and No. 18K18743. NT, HS, NT were supported by JST-MIRAI program, Grant No. JPMJMI18A3, Japan. TM acknowledges support from JST Mirai JPMJMI19A1.

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Fig. 1: X-ray powder diffraction pattern of prepared YbCo$_2$ compound crystalized in cubic Laves phase MgCu$_2$-type.

Fig. 2: Temperature dependent of magnetic susceptibility measured at 1 T. Black solid line represents Curie-Weiss fit. Inset graph shows magnetization curves measured at different temperatures.

Fig. 3: Derivative of magnetization curves showing local maximum at low temperatures.
Fig. 4: Temperature dependence of magnetization and its derivative measured at several different magnetic fields.

Fig. 5: Temperature dependent of electrical resistivity. Inset graph shows resistivity curve in low temperature.

Fig. 6: Temperature dependent of electrical resistivity measured at different magnetic fields (a) where arrow marks a transition-like anomaly. Derivative of each curve in low temperature is plotted in section (b).
Fig. 7: Fit of temperature dependence of electrical resistivity measured at different magnetic fields.

Fig. 8: Specific heat curves measured on YbCo$_2$ (red squares) and non-magnetic analog YNi$_2$ (green squares). Inset shows detail of YbCo$_2$ specific heat curve in low temperature.
Fig. 9: Magnetic specific heat of YbCo$_2$ (red squares) and fitting of Schottky contribution (blue line), calculation of Kondo contribution into specific heat by Coqblin-Schriefer model (green and black lines)$^{28}$.

Fig. 10: Low temperature part of specific heat over temperature curves measured at different magnetic fields.
Fig. 11: Constructed phase diagram of magnetic transition at YbCo₂.