Pressure-induced ferromagnetism with strong Ising-type anisotropy in $YbCu_2Si_2$

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We report dc magnetic measurements on $YbCu_2Si_2$ at pressures above 10 GPa using a miniature ceramic anvil cell. $YbCu_2Si_2$ shows a pressure-induced transition from the nonmagnetic to a magnetic phase at 8 GPa. We find a spontaneous dc magnetization in the pressure-induced phase above 9.4 GPa. The pressure dependence of the ferromagnetic transition temperature T_C and the spontaneous magnetic moment μ_0 at 2.0 K have been determined. The value of μ_0 in the present macroscopic measurement is less than half of that determined via Mössbauer experiments. The difference may be attributed to a spatial phase separation between the ferromagnetic and paramagnetic phases. This separation suggests that the pressure-induced phase boundary between the paramagnetic and ferromagnetic states is of first order. Further, we have studied the magnetic anisotropy in the pressure-induced ferromagnetic state. The effect of pressure on the magnetization with magnetic field along the magnetic easy *c* axis is much larger than for field along the hard *a* axis in the tetragonal structure. The pressure-induced phase has strong Ising-type uniaxial anisotropy, consistent with the two crystal electric field models proposed for $YbCu_2Si_2$.

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I. INTRODUCTION

In recent years there has been growing interest in strongly correlated electron systems of rare earth and actinide compounds located at or close to a magnetic quantum critical point (QCP) [1]. The electronic state of such systems can often be tuned with pressure or magnetic fields. Unconventional superconductivity and non-Fermi-liquid behavior have been observed near pressure-induced magnetic to nonmagnetic phase boundaries in many cerium compounds such as $CeIn_3$ [2]. The novel physical phenomena have been studied from the viewpoint of the quantum criticality. Such phenomena might be expected in ytterbium compounds since Yb is considered to be a "hole" equivalent of Ce. Indeed, anomalous physical properties have been reported and extensively studied in $YbRh_2Si_2$ and β-$YbAlB_4$ [3,4].

Application of pressure tends to drive the Yb ion from nonmagnetic Yb^{2+} ($4f^{14}$) to magnetic Yb^{3+} ($4f^{13}$) states. A magnetic ordered state is stabilized at higher pressures. A pressure-induced magnetic phase has been reported in a number of Yb compounds. In most cases the pressure-induced magnetic phase has been detected via ac magnetic susceptibility measurements. There have been few studies of detailed magnetic properties of a pressure-induced phase using dc magnetization measurements. This is due to the common experimental constraint that the maximum pressure is only 1.5 GPa for the most commonly used piston-cylinder-type cell in a commercial superconducting quantum interference device (SQUID) [5].

Recently, we have developed a miniature ceramic anvil cell (mCAC) for magnetic measurements at pressures above 10 GPa with the use of the SQUID magnetometer [6–8]. Owing to the simplicity of the cell structure, the mCAC can detect the ferromagnetic ordered state whose spontaneous magnetic moment is significantly less than $1.0\mu_B$ per magnetic ion. The cell enables us to make a quantitative study of the pressure-induced phases in Yb compounds. We report here a study of the anisotropic magnetic properties of the pressure-induced phase in $YbCu_2Si_2$.

$YbCu_2Si_2$ crystallizes in the tetragonal $ThCr_2Si_2$ structure. This is a paramagnetic compound with a moderately high value of the linear specific heat coefficient $\gamma \simeq$ 135 mJ K^{-2} mol^{-1} [9,10]. Previous high-pressure studies suggested a pressure-induced, possibly ferromagnetic, ordered state above 8 GPa from ac magnetic susceptibility measurements and Mössbauer experiments [11–14]. It is therefore important to detect the ferromagnetic component from dc magnetic measurements at high pressure. In this study we have measured the magnetization of $YbCu_2Si_2$ with our mCAC.

II. EXPERIMENT

Single crystals of $YbCu_2Si_2$ were grown from Sn flux [9,10]. We have used our miniature ceramic anvil high-pressure cell mCAC with 0.6 mm culet anvils [6–8]. The Cu-Be gasket was preindented to 0.08 mm from an initial thickness of 0.30 mm. The diameter of the sample space in the gasket was 0.20 mm. To study the anisotropy of the magnetic properties in $YbCu_2Si_2$, two single crystals were measured with a magnetic field applied parallel to the magnetic easy *c* axis (the [001] direction) and the hard *a* axis ([100] direction) in the tetragonal crystal structure. The sizes of the single crystal samples were $0.11 \times 0.09 \times 0.03$ mm^3 and $0.10 \times 0.09 \times 0.02$ mm^3 for magnetic measurements with a magnetic field along the *c* axis and the *a* axis, respectively. The sample and a Pb pressure sensor were placed in the sample space filled with glycerin as the pressure-transmitting medium [15]. The pressure values at low temperatures were determined by the pressure dependence of the superconducting transition temperature of Pb [16–18]. The pressure medium glycerin solidifies at 5 GPa at room temperature. The pressure

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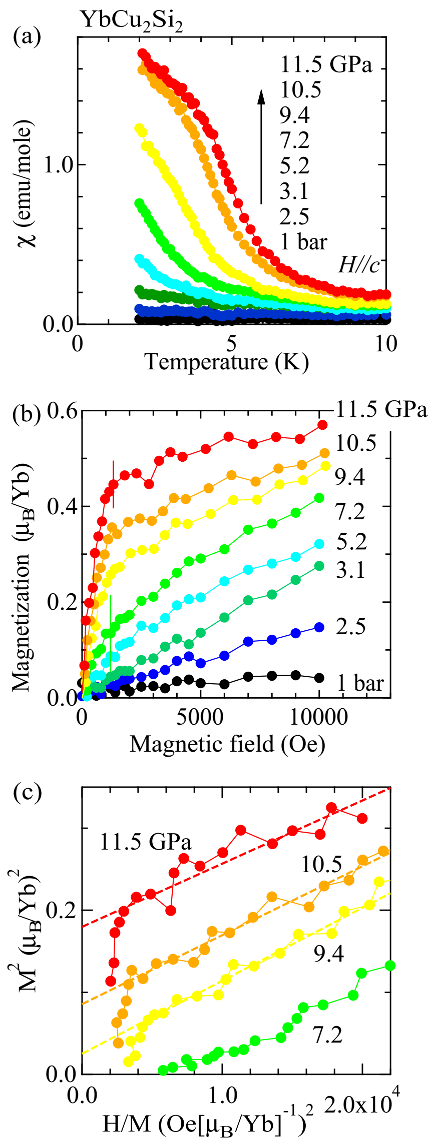

FIG. 1. (Color online) (a) Temperature dependence of the magnetic susceptibility χ under a magnetic field of 1 kOe applied along the magnetic easy c axis ($H||[001]$). (b) Magnetic field dependence of the magnetization and (c) Arrott plots of the magnetization measured at 2.0 K and at 1 bar, 2.5, 3.1, 5.2, 7.2, 9.4, 10.5, and 11.5 GPa.

inhomogeneity was estimated as $\Delta P \sim 1$ GPa above 10 GPa. The demagnetization effect needs to be taken into account in the pressure-induced ferromagnetic state. The internal field values $H_{\rm int}$ were determined by subtracting the demagnetizing field given by $H_{\rm int} = H_{\rm appl} - DM$. Here, $H_{\rm appl}$ is the external magnetic field and D is the demagnetizing factor. Error bars in Fig. 1(b) indicate possible errors in the estimation of the magnetization.

III. RESULTS AND DISCUSSIONS

Figure 1 shows the temperature dependence of the magnetic susceptibility χ in a magnetic field of 1 kOe applied along the magnetic easy c axis ($H||[001]$) [Fig. 1(a)] and the magnetic field dependence of the magnetization measured at 2.0 K and at 1 bar, 2.5, 3.1, 5.2, 7.2, 9.4, 10.5, and 11.5 GPa [Fig. 1(b)]. At 1 bar, χ shows an almost temperature-independent value of $\chi = 0.03$ emu/mol below 10 K and the magnetization increases linearly with increasing magnetic field, consistent with the previous study [9]. Application of pressure above 5 GPa induces a low-temperature upturn in χ and a nonlinear increase of the magnetization in low fields. At 9.4, 10.5, and 11.5 GPa, the magnetization shows typical ferromagnetic behavior with the magnetic susceptibility, diverging at low temperatures. These results indicate that the pressure-induced magnetic transition in $YbCu_2Si_2$ is ferromagnetic.

The spontaneous magnetic moment μ_0 is determined above 9.4 GPa from the Arrott plot shown in Fig. 1(c). The values of μ_0 at 2.0 K are estimated as 0.16 ± 0.08, 0.30 ± 0.08, and $0.42 \pm 0.05 \mu_B$/Yb at 9.4, 10.2, and 11.5 GPa, respectively. The ferromagnetic transition temperatures T_C at 9.4, 10.5, and 11.5 GPa are estimated as 3.5 ± 0.5, 4.3 ± 0.5, and 4.7 ± 0.5 K, respectively, from the peak position in the temperature derivative of the magnetic susceptibility $\partial\chi/\partial T$.

Figure 2 shows the pressure dependences of the ferromagnetic transition temperature T_C [Fig. 2(a)] and the spontaneous magnetic moment μ_0 at 2.0 K in $YbCu_2Si_2$ [Fig. 2(b)]. A ferromagnetic transition was not observed down to 2.0 K at 8.8 GPa (data not shown). The transition may occur below 2.0 K. The critical pressure P_c for the ferromagnetic state may be located between 8.0 and 8.5 GPa. Fernandez-Pañella *et al.* reported that the pressure effect on T_C depends largely

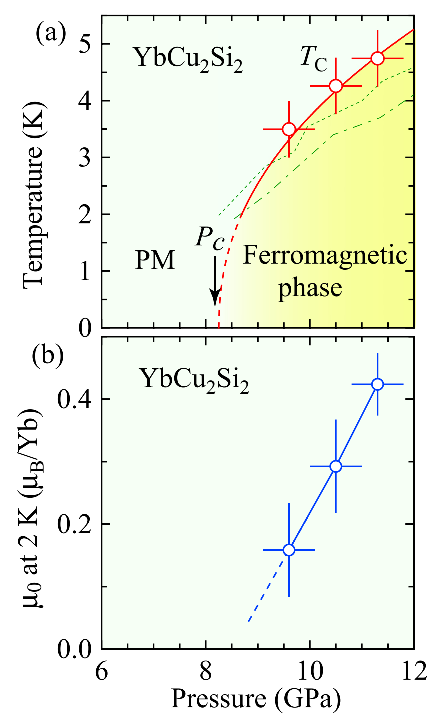

FIG. 2. (Color online) (a) Temperature-pressure phase diagram of $YbCu_2Si_2$. Circles indicate the ferromagnetic transition temperature T_C. Dotted and dashed-dotted lines indicate the pressure dependences of T_C for the sample with a residual resistance ratio of RRR = 200 in the previous study (Ref. [14]). The former and the latter lines were determined by the ac magnetic susceptibility measurement and the ac calorimetry, respectively. (b) Pressure dependence of the spontaneous magnetic moment μ_0 determined at 2.0 K.

on the sample quality [14]. The present pressure dependence of T_C is consistent with those for samples with a similar quality (RRR = 200) as in the previous study, shown as dotted and dashed-dotted lines in Fig. 2(a). The former and the latter lines were determined by the ac magnetic susceptibility measurement and ac calorimetry, respectively.

It has been established that, above critical pressure P_c, the transition to the ferromagnetic phase in $YbCu_2Si_2$ is of first order [13,14]. Indeed, the ac magnetic susceptibility measurement showed a sudden appearance of a ferromagnetic transition above 1 K [14]. However, no sharp anomaly at T_C is observed in the temperature dependence of the magnetization at any pressure above 9.4 GPa, indicating a second-order phase transition. We suggest that the ferromagnetic transition changes from the first- to the second-order phase transition at a somewhat higher pressure than P_c in $YbCu_2Si_2$. P_c may be a weakly first-order critical point. This may be a reason for the absence of non-Fermi-liquid behavior in the resistivity ρ. It shows a typical Fermi-liquid behavior $\rho = \rho_0 + AT^2$ down to 30 mK around P_c, where ρ_0 is the residual resistivity [11]. The value of A increases continuously as a function of the pressure but it does not show a divergent behavior around P_c. Several ferromagnets such as $ZrZn_2$ [19], $Co(S_{1-x}Se_x)_2$ [20], MnSi [21], and UGe_2 (Ref. [22]) have a tricritical point where the paramagnet to ferromagnet transition changes from a second-order to a first-order phase transition when driven toward the QCP by applying external pressure or chemical pressure. This seems to be a general property of ferromagnets, as has been theoretically discussed [23].

We discuss the pressure-induced ferromagnetism in $YbCu_2Si_2$ from two points of view. There are two crystal electric field (CEF) models (I and I′) proposed for $YbCu_2Si_2$ in previous studies [9]. The values of the magnetic moment expected from the doublet ground state are 2.70 and $2.29\mu_B$/Yb for the CEF models I and I′, respectively. We compare the values in the CEF models with that determined in the Mössbauer experiment (i) and that determined in the present macroscopic measurement (ii).

(i) The values of the magnetic moment in the CEF models are more than two times larger than that ($1.25\mu_B$/Yb) determined with the Mössbauer experiment at 8.9 GPa at 1.8 K [12]. The reduced magnetic moment in the Mössbauer experiment may be due to the Kondo effect. Resonant inelastic x-ray scattering measurements showed that the value of the Yb valence is 2.88 at 7 K near P_c [24]. Thus, ferromagnetism appears in the mixed valence state in $YbCu_2Si_2$. Contrary to cerium compounds, a magnetic ordering can appear in the intermediate valence state ($n_{4f} \ll 1$) of the Yb systems, where n_{4f} is the occupation number of the $4f$ level [25–27]. Differences in the magnetic properties between the Ce and Yb systems arise from differing hierarchies of the energy scales of the Kondo temperature T_K, the $4f$ bandwidth Δ_{4f}, and the splitting energy between ground and first excited states in the CEF levels Δ_{CEF} [25–27]. T_K of Yb systems could be smaller or comparable to Δ_{CEF} because of the smaller Δ_{4f} in Yb systems than that in cerium systems. In $YbCu_2Si_2$, the electrical resistivity under high pressure suggests that T_K is less than 50 K at around P_c [11]. The value of T_K is lower than that of Δ_{CEF} in models I and I′ [9]. The linear specific heat coefficient γ is estimated as $\gamma \sim 1$ J/mol K^2 at P_c from the coefficient A of the T^2 term in the resistivity with the Kadowaki-Woods relation [11,28]. The pressure-induced phase in $YbCu_2Si_2$ is a ferromagnetic heavy fermion system with an intermediate valence of the Yb ion. This is opposed to Ce systems where the magnetic ordering or heavy fermium states are usually restricted to the trivalent configuration ($n_{4f} \sim 1.0$) [25–27].

(ii) In the present macroscopic magnetic measurement, the spontaneous magnetic moment μ_0 at 2.0 K and 9.4 GPa is less than half of that with the Mössbauer experiment [12]. This difference may be due to a spatial phase separation between the paramagnetic and ferromagnetic states suggested in the Mössbauer spectrum. The value of μ_0 increases with increasing pressure. The continuous change in μ_0 around the critical pressure P_c is difficult to understand because the pressure-induced change is of first order [13,14]. We point out two possibilities. One is that the volume fraction of the ferromagnetic phase increases as a function of pressure and the other is that the pressure change of μ_0 reflects the increase of the Yb valence above P_c as seen in the resonant inelastic x-ray scattering measurement [24]. The phase separation suggests a first-order phase boundary between the paramagnetic and the ferromagnetic phases.

Figure 3 shows the temperature dependence of the magnetic susceptibility χ in a magnetic field of 1 kOe applied along the magnetic hard a axis ($H||[100]$) [Fig. 3(a)] and the magnetic field dependence of the magnetization measured at 2.0 K and at 1 bar, 5.0, 9.0, 11.0, and 12.2 GPa [Fig. 3(b)]. Compared with the magnetization data for $H||c$, the pressure effect on the magnetization for $H||a$ is significantly smaller. The value of χ at

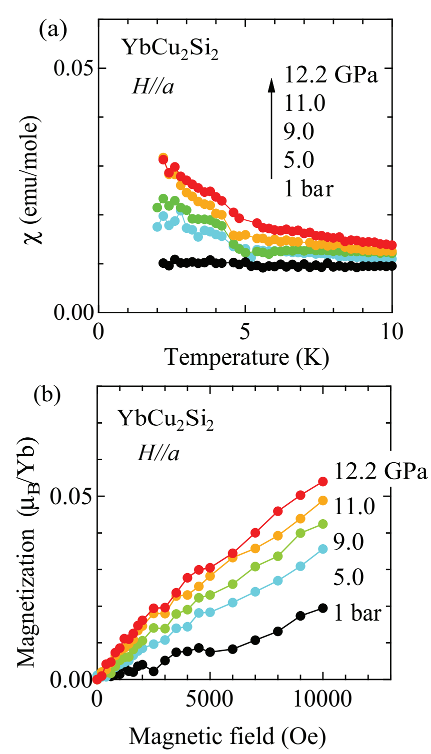

FIG. 3. (Color online) (a) Temperature dependence of the magnetic susceptibility χ under a magnetic field of 1 kOe applied along the magnetic hard a axis ($H||[100]$) and (b) a magnetic field dependence of the magnetization measured at 2.0 K and at 1 bar, 5.0, 9.0, 11.0, and 12.2 GPa.

2.0 K is increased from 0.01 emu/mol at 1 bar to 0.03 emu/mol at 12.2 GPa. The magnetization curve does not show a ferromagnetic behavior at higher pressures. The magnetic-field-induced moment at 10 kOe is $0.038\mu_B$/Yb at 12.2 GPa, one order of magnitude smaller than that ($0.57\mu_B$/Yb) with a magnetic field applied along the easy c axis at 11.5 GPa. The pressure-induced ferromagnetic phase has strong uniaxial anisotropy. The Ising character of the magnetic property is suggested from the two CEF models proposed for $YbCu_2Si_2$ [9].

The Ising-type magnetic fluctuation can induce the spin-triplet p-wave superconductivity around the ferromagnetic QCP [1]. A motivation for the previous high-pressure studies on $YbCu_2Si_2$ was to search for the superconductivity around P_c. However, the superconductivity has not been found in resistivity measurements down to 30 mK [11]. Theoretically, the superconducting transition temperature for spin-triplet p-wave pairing around the ferromagnetic QCP is largely lower than that of the spin-singlet d-wave superconductivity around the antiferromagnetic QCP [29]. In Ce systems, $CeIn_3$ and $CeRhIn_5$ exhibit superconductivity under high pressure where T_{sc} attains maximum values of 0.2 and 2.2 K, respectively [2,30]. In uranium systems, the superconductivity appears in the ferromagnetic state of UGe_2 [31]. The value of T_{sc} is 0.8 K at 1.2 GPa. URhGe and UCoGe show the superconducting transition at $T_{sc} = 0.2$ and 0.7 K, respectively, at ambient pressure [32,33]. The characteristic temperature of the electronic state in Yb systems is lower than those in Ce and U systems due to the smaller bandwidth of the $4f$ band, as mentioned before. If the superconductivity existed in $YbCu_2Si_2$, the transition temperature would be very low. This may be a reason why the heavy fermion superconductivity of the $4f$ electrons is elusive in Yb systems. Also, spatial phase separation in $YbCu_2Si_2$ may be harmful for the appearance of superconductivity.

The present study shows convincing evidence of ferromagnetism in the pressure-induced phase of $YbCu_2Si_2$ from dc magnetization measurements. Ferromagnetism has been found in a number of Yb compounds such as YbRhSb, $YbInNi_4$, and YbNiSn at ambient pressure [34–36], and $YbInCu_4$ and $YbIr_2Si_2$ at high pressure [37,38]. On the other hand, there are only a few cerium-based compounds such as $CeRh_3B_2$ and CeAg which show a ferromagnetic ground state [39,40]. The origin of this difference is an interesting question. The hierarchies of the energy scales of T_K, Δ_{4f}, and Δ_{CEF} in the Ce and Yb systems are different, as mentioned before [25–27]. This leads to the larger change in valence of the Yb ions from the nonmagnetic 2+ to magnetic 3+ in real lattices, as compared with that of the Ce ions. The resonant inelastic x-ray scattering experiment shows a wider valence change in $YbCu_2Si_2$, as compared with that in its Ce counterpart, $CeCu_2Si_2$ [24]. The valence transition or instability of the Yb ion has been detected by x-ray absorption or emission spectroscopy in $YbAgCu_4$ [41], $YbInCu_4$ [42], and $YbCu_{5-x}Al_x$ [43]. Recently, new aspects in strongly correlated electron systems originating from valence fluctuation of the rare earth ion have been theoretically discussed [44,45]. Anomalous physical properties in β-$YbAlB_4$ and $YbRh_2Si_2$ have been reconsidered from this point of view [45]. The theoretical study also shows a simultaneous divergence of the valence susceptibility and the uniform spin susceptibility at the quantum critical point of the valence transition under a magnetic field. This strengthens the ferromagnetic tendency in Yb systems under finite magnetic field. Careful future theoretical study is necessary for a realization of ferromagnetism under a zero magnetic field [46]. From the experimental point of view, comprehensive studies on the Yb systems should be done to reveal the valence state of the Yb ions in the wide temperature, magnetic field, and pressure regions.

IV. CONCLUSION

In conclusion, dc magnetic measurements have been done to study the magnetic property of the pressure-induced phase in $YbCu_2Si_2$ with a miniature ceramic anvil high-pressure cell. The ferromagnetic ordered state is confirmed from the observation of dc spontaneous magnetization. The pressure dependences of the ferromagnetic transition temperature T_C and the spontaneous magnetic moment μ_0 at 2.0 K have been determined. The value of μ_0 in the present macroscopic measurement is less than half of that determined via Mössbauer experiments, which may be attributed to a spatial phase separation between the ferromagnetic and paramagnetic phases. Peculiar features in the pressure-induced ferromagnetic state are discussed in comparison with cerium compounds. The effect of pressure on the magnetization with a magnetic field along the magnetic easy c axis is much larger than for a field along the hard a axis in the tetragonal structure. The pressure-induced phase in $YbCu_2Si_2$ has strong Ising-type uniaxial anisotropy, consistent with the two crystal electric field (CEF) models proposed for $YbCu_2Si_2$.

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