

Geometrical frustration versus magnetic order in the heavy-fermion antiferromagnet YbAgGe under high pressure

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We have investigated the effect of pressure on the magnetic and structural properties of the frustrated heavy-fermion antiferromagnet YbAgGe in an extended pressure range up to 20 GPa using electrical resistivity and x-ray diffraction, respectively. We find that with increasing pressure, the magnetic ordering temperature (T_m) first increases rapidly, passes through a maximum ($T_m=5.4$ K at 6.8 GPa), and then drops toward zero for p above 16 GPa, while the structure remains unchanged up to about 20 GPa. We attribute the anomalous pressure dependence of T_m to a complex interplay between the Kondo effect, Ruderman-Kittel-Kasuya-Yosida interactions and the geometrical frustration of the magnetic interactions of the Yb moments in the Kagomé-latticelike structure of YbAgGe. We further discuss the competition between geometrical frustration and long-range magnetic order together with the possible occurrence of a pressure-induced second quantum critical point in YbAgGe.

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I. INTRODUCTION

The investigation of strongly correlated f -electron systems such as Ce or Yb heavy-fermion compounds near a magnetic quantum-critical point (QCP) has recently been the subject of continuous interest due to the observation of unusual ground states. In such systems, the crossover from magnetically ordered (MO) to a nonmagnetic (NM) state or vice versa is mainly driven by the hybridization strength J between the f and conduction electrons which determines the ratio between the characteristic energies for the Kondo effect (T_K) and the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction (T_{RKKY}).¹ As a result of the competition between low energy-scale interactions, these materials exhibit complex phase diagrams and nontrivial ground states. What is particularly interesting is that J can be experimentally tuned by a nonthermal control parameter such as chemical substitution, magnetic field, or external pressure and thereby allows one to investigate the nature of the ground state close to the borderline between the MO and NM states. Known examples for pressure tuning are non-Fermi-liquid behavior in CeCu_{6-x}Au_x (Ref. 2) and Ce₇Ni₃ (Ref. 3) pressure-induced superconductivity in CePd₂Si₂,⁴ and an unusual low moment state in YbRh₂Si₂ under pressure.⁵

In this respect, the study of the geometrically frustrated MO heavy-fermion compound YbAgGe, located near to a QCP is of particular interest due to the fact that the frustrated magnetic interactions compete with the Kondo effect.⁶⁻⁹ YbAgGe crystallizes in the hexagonal ZrNiAl-type structure,¹⁰ where the Yb moments are aligned in the basal plane of the structure, forming a hexagonal array similar to a Kagomé lattice, which results in frustrated antiferromagnetic interactions. In addition, the crystal electric field at the Yb site induces a strong single-ion anisotropy. This magnetic frustration leads to anomalous magnetic properties: YbAgGe

undergoes two successive magnetic transitions at $T_{M1}=0.8$ K and $T_{M2}=0.65$ K (Ref. 9) and exhibits strong spin fluctuations at temperatures ($T=10$ K), much larger than those of antiferromagnetic ordering. The latter has been reported from inelastic neutron-scattering experiments on YbAgGe.¹¹ Heavy-fermion behavior is also reported for YbAgGe (electronic specific-heat coefficient $\gamma \approx 200$ mJ/K² mol).⁹ These anomalous low-temperature properties together with the relatively high Kondo temperature ($T_K=25$ K) justify the assumption that YbAgGe is located near a magnetic QCP. Thus, YbAgGe is an excellent candidate for the investigation of the complex interplay between different low energy-scale interactions under high pressure.

Unlike Ce-based compounds ($4f^1, \text{Ce}^{3+}$) where the applied pressure (which causes the volume decrease) suppresses the MO state, in Yb-based compounds ($4f^{13}, \text{Yb}^{3+}$) it stabilizes (induces) the MO state.^{5,12-14} Indeed, recent high-pressure studies on YbAgGe by some of the authors (electrical resistivity and specific heat) up to about 3 GPa revealed significant change in the magnetic state:^{9,15} for $p=0.5$ GPa, the two successive magnetic transitions at T_{M1} and T_{M2} merge into one at $T_{M3}=0.8$ K, and remains nearly constant up to about 1.5 GPa. It then rapidly increases to about 1.7 K at $p=3.2$ GPa.¹⁵ This unusual behavior has been suggested to be due to a crossover to a stable moment regime, in which spin fluctuations and the associated magnetic frustrations are suppressed with increasing pressure above 1.5 GPa. However, since the Yb magnetic moments in this pressure range are still geometrically frustrated in the hexagonal structure, an open question is to what extent geometrical frustration affects the long-range magnetic order in the MO state upon further increasing pressure. In fact magnetic frustration in a Heisenberg-type spin system can hinder long-range magnetic order of the magnetic moments down to $T=0$ K.¹⁶⁻¹⁹ In or-

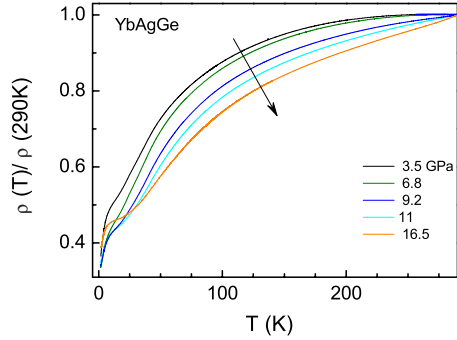


FIG. 1. (Color online) Temperature dependence of the electrical resistivity normalized at 290 K for different selected pressures. The arrow indicates the evolution from low to high pressure.

der to provide an answer to this interesting question, we have investigated the effect of pressure on the magnetic and structural properties of YbAgGe in an extended pressure range up to about 20 GPa.

II. EXPERIMENT

High-quality single-crystalline YbAgGe samples were prepared according to the method described in Ref. 8. Samples checked by x-ray diffraction were single phase [hexagonal ZrNiAl-type structure, space group $P6_2m$ (Ref. 10)]. The pressure dependence of the electrical resistance between 1.5 and 300 K and up to about 17 GPa has been measured using the four-point technique in a diamond-anvil cell (DAC) made from a special Ti alloy which ensures thermal stability against temperature variations (constant pressure). The metallic gasket (Inconel 750) of the DAC was insulated from the four leads by a thin layer of epoxy and Al_2O_3 mixture, serving also as pressure transmitting medium. The resulting sample chamber had a diameter and a height of 100 and 50 μm , respectively. The distance of the leads determining the voltage drop on the sample was about 50 μm . The pressure gradient within the pressure cavity was about 5–7 %. Pressure was measured by the ruby luminescence method.^{20,21} Data were taken in a 4He bath cryostat using dc current and by measuring the voltage drop for both polarities. The measurements of the lattice parameters as a function of pressure were performed by energy dispersive x-ray diffraction (EDXRD) at the Hamburger Synchrotronstrahlungslabor (HASYLAB), beam line F3 using the DAC (same type) technique up to about 20 GPa at 300 K. Liquid Nitrogen was used as pressure transmission medium and the pressure was determined by the ruby luminescence as in electrical resistivity measurements.

III. RESULTS

Figure 1 displays the temperature dependence of the electrical resistivity $\rho(T, p)$ of YbAgGe between 1.5 and 300 K at different pressures up to about 16.5 GPa. In all cases, $\rho(T, p)$ decreases slowly with decreasing temperature, showing a convex curvature, followed by a rapid decrease well below about 10 K. The anomalous change in $\rho(T, p)$ below

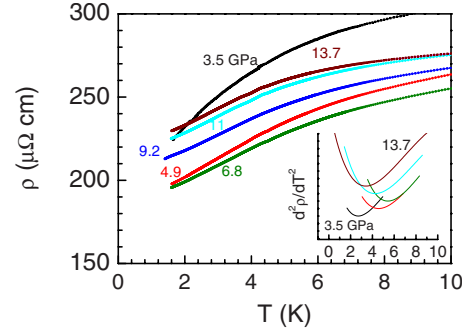


FIG. 2. (Color online) Low-temperature (≤ 10 K) part of the resistivity at selected pressures. The inset shows the second derivatives of $\rho(T, p)$ curves for different pressures.

300 K is most probably due to an incoherent Kondo scattering of the conduction electrons at the first excited crystal-field level. Indeed, recent inelastic neutron-scattering experiments on YbAgGe reveal a single excitation at about 12 meV (≈ 140 K).²²

At temperatures below 10 K (see Fig. 2), $\rho(T, p)$ decreases more rapidly signaling the onset of magnetic ordering as usually observed in many magnetically ordered Yb-based heavy-fermion compounds.^{12,13,23} Interestingly, one first observes a decrease of $\rho(T, p)$ with increasing pressure up to 6.8 GPa and then an increase for higher pressures up to 16.5 GPa.

To obtain values of the residual resistivity ρ_0 at different pressures up to 16.5 GPa, we have performed polynomial fits to the data and extrapolated to zero. These values of ρ_0 as a function of pressure are plotted in Fig. 3(a). It is evident that with increasing pressure, ρ_0 remains nearly constant up to about 6.8 GPa and then abruptly increases up to 16.5 GPa. This clear increase in the value ρ_0 reflects a change of the spin-disorder scattering below and above 6.8 GPa, which should be related to a pressure-induced change in the nature of magnetic order in YbAgGe above 6.8 GPa. The change in the magnetic ordering temperature (T_m) with increasing pressure is deduced from the minimum in the second derivative in $d^2\rho/dT^2$. These are shown in the inset of Fig. 2 for some selected pressures. Figure 3(b) displays the pressure dependence of T_m together with those previously measured in the low-pressure range up to 3 GPa.¹⁵ As shown in Fig. 3(b), T_m sharply increases with increasing pressure from about 1.5 K to a maximum value of about 5.4 K at about 6.8 GPa and then drops to about 2.2 K at 13.8 GPa. We find no minimum in $d^2\rho/dT^2$ for the data collected at 16.5 GPa. An extrapolation of the measured $T_m(p)$ in the Fig. 3(b) indicates that magnetic ordering would disappear ($T_m \rightarrow 0$) at a critical pressure of about 16 GPa. We notice from the comparison of $T_m(p)$ and ρ_0 [see Figs. 3(a) and 3(b)] that while for $p=0$, the increase in $T_m(p)$ is not associated with a change in $\rho_0(p)$, the suppression of T_m above 6.8 GPa corresponds to an increase in $\rho_0(p)$. This mutual relationship between the pressure-induced change in T_m above 6.8 GPa and that of the spin-disorder scattering supports our suggestion of a pressure-induced change in the nature of magnetic order in YbAgGe above 6.8 GPa.

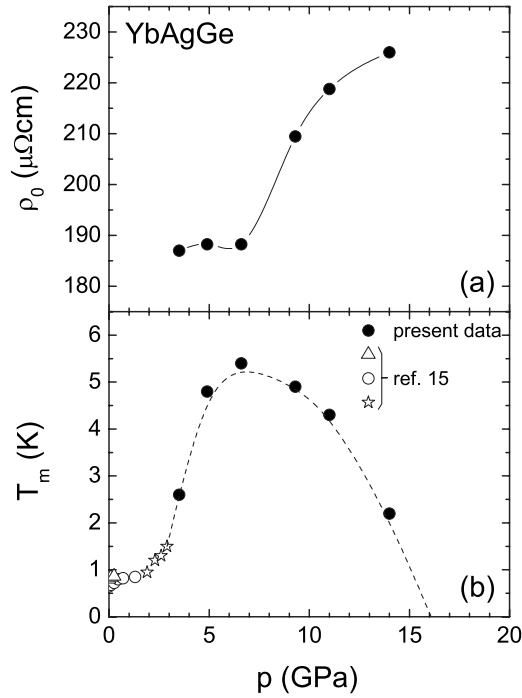


FIG. 3. (a) Change in the residual resistivity (ρ_0) as a function of pressure as obtained from the data shown in Fig. 2. The line through the data points is guide to the eye; (b) pressure dependence of the magnetic ordering temperature T_m up to 13.8 GPa [closed circles (●) represent the present data]. Experimental points at low pressures up to 3 GPa (Δ , \circ , \star) are taken from Ref. 14. Dashed line through data points is only guide to the eye.

To exclude that the observed anomalous pressure dependence of T_m is related to a corresponding structural changes (e.g., phase transition), we now consider the effect of pressure on the lattice structure of YbAgGe as obtained from our high-pressure EDXRD measurements. Figure 4 shows some selected typical EDXRD patterns collected at 300 K up to 20 GPa. The space group of the hexagonal ZrNiAl-type

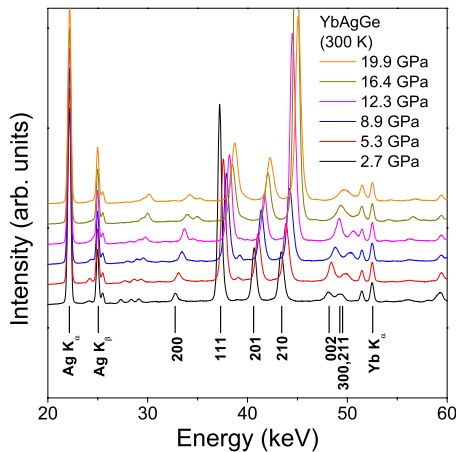


FIG. 4. (Color online) X-ray diffraction pattern of YbAgGe at room temperature for selected pressures up to ~ 20 GPa. Spectra were taken at beamline F3, HASYLAB, Hamburg.

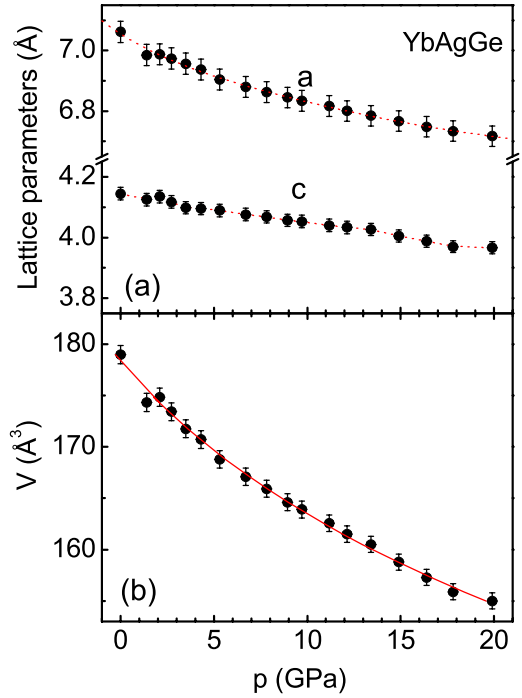


FIG. 5. (Color online) (a) Pressure variation in the lattice parameters a and c obtained from the analysis of EDXRD pattern. The dotted line through data points are only a guide to the eye; (b) pressure dependence of the unit cell volume of the YbAgGe V up to 20 GPa and at room temperature. The solid line through the data in is a fit using Birch's equation of state.

structure could identify all peaks. From the analysis of the data, we obtain a smooth variation in the lattice parameters (a and c) and the volume of the unit cell as shown in Fig. 5. This clearly indicates that no structural phase transition occurs up to 20 GPa. A fit of the data using Birch's equation of state²⁴ results in values of the modulus $B_0 = 82 \pm 7$ GPa and its pressure derivative $B'_0 = 7 \pm 1$.

IV. DISCUSSION

According to the x-ray data, the unusual pressure dependence of T_m in YbAgGe is not connected with a structural phase transition. An important consequence of this finding is that the geometrically frustrated hexagonal (ZrNiAl) structure remains unchanged up to 20 GPa. This implies that the geometrically frustrated magnetic interactions should be taken into account to explain the anomalous pressure dependence of T_m and its rapid decrease for pressures above 6.8 GPa. In this context, we discuss in the following the unusual change in T_m with pressure in YbAgGe. As we mentioned above, at ambient pressure, the frustrated antiferromagnet YbAgGe is located very close to a magnetic QCP. Thus, its ground state is not only determined by the balance of the Kondo effect, RKKY interactions and crystal field anisotropy, but also by the interplay between those energy scales with the geometrical frustration of the magnetic interactions. Furthermore, the system exhibits strong spin fluctuations at

temperatures much higher than T_{M1} and T_{M2} . It is anticipated that such fluctuations lead to an additional enhancement of magnetic frustration, in particular, near the QCP. Thus, magnetic frustrations and the strength of spin fluctuations are intimately related and cannot be treated independently.

Since in Yb-based magnetically ordered compounds, external pressure gradually moves the system away from the QCP and thereby stabilizes the long-range magnetic order, the pressure dependence of T_m is determined by the relative strength of all types of competing energy scales with increasing pressure. Increasing the pressure up to 6.8 GPa, both the Kondo effect and spin fluctuations (and the associated quasiparticles) will be gradually suppressed, while RKKY interactions increase. These together with the expected increase in the crystal-field anisotropy,^{25,26} which causes a corresponding increase in the magnetocrystalline anisotropy leads to the observed increase in T_m with pressure. Such an increase in T_m is expected to be connected with a gradual restoration of the localized moments of Yb. Indeed, very recent high-pressure high-field-magnetization measurements on single crystals of YbAgGe at pressures up to $p=2.2$ GPa show a significant increase in the Yb magnetic moment of about 20% as well as an increase in the magnetocrystalline anisotropy along the a axis.²⁷ These findings support our explanation of the increase in T_m with increasing pressure for p up to 6.8 GPa.

Thus, at $p=6.8$ GPa, where T_m reaches its maximum value ($T_m=5.4$ K), the system is expected to exhibit localized Yb magnetic moments. In this connection, it has to be mentioned that the pressure-induced suppression of spin-fluctuations results in a corresponding decrease in the associated magnetic frustrations. However, as the local Yb moments are recovered with increasing pressure, geometrical frustrations become more significant and are expected to play a dominating role by further increasing pressure above 6.8 GPa. As shown in Fig. 4, for pressures above 7 GPa T_m rapidly decreases toward zero, suggesting the disappearance of magnetic order for pressure at around 16 GPa as a second QCP. The impact of geometrical frustration of magnetic exchange interactions on the nature of magnetically ordered state is demonstrated by our findings [see Figs. 3(a) and 3(b)] that the pressure-induced change in T_m in the whole pressure range is associated with a corresponding change of the spin-disorder scattering: the increase in T_m up to 6.8 GPa is not connected to a change in ρ_0 , thus indicating a spin-disorder scattering, expected for ordered local magnetic moments. In contrast, the decrease in T_m above 6.8 GPa is associated with an increase in the spin-disorder scattering, which suggests a gradual formation of magnetically frustrated ground state. In this context, we would like to mention that in principle a decrease in $T_m(p)$ might be due to a pressure-induced enhancement of atomic disorder as recently theoretically reported.²⁸ According to these calculations the transition from the conventional ordered (CO) phase to a Griffiths paramagnetic phase with increasing disorder takes place through the formation of a cluster glass (CG) phase. The transition from the CO to CG phase can be either continuous or abrupt. However, since atomic disorder, if present, would be expected to gradually increase with increasing pressure such a theoretical scenario cannot explain the observed pres-

sure dependence of T_m in the whole pressure range, i.e., between ambient pressure and 16.5 GPa through a maximum value around 7 GPa. We, thus, would like to attribute the observed rapid decrease in T_m for $p=6.8$ GPa and the disappearance of long-range magnetic ordering to a pressure-induced modification of the frustrated magnetic interactions in the Kagomé-type lattice structure of YbAgGe as discussed below.

This explanation is based on the assumption that the Yb-magnetic moments in the basal plane can be treated as a quasi-two-dimensional Heisenberg spin, $S=1/2$ system, forming a frustrated Shastry-Sutherland lattice.^{16–19} This is supported by the inelastic neutron-scattering results which reveal the absence of \mathbf{q} dependence of the magnetic susceptibility within the basal plane of the hexagonal lattice of YbAgGe, clearly indicating in-plane frustration.¹¹ In such a frustrated lattice, the ground state is shown to be determined by the ratio of the exchange-coupling constants (J'/J) of the nearest-neighbor (J) and next-nearest-neighbor (J') atoms. Accordingly, a quantum phase transition occurs at $J'/J=0.7–0.9$ from the magnetically ordered to a nonmagnetic state. The latter is favorable for values of J'/J less than such critical values. Following such a scenario, we can explain the decrease in T_m with increasing pressure above 7 GPa toward zero by a pressure-induced decrease in J'/J . We note, that the critical ratio may be different for different systems and has to be calculated in the specific case of YbAgGe. However, for a qualitative explanation, it is only important to show that a critical value of J'/J can lead to a QCP. This qualitative interpretation of a pressure-dependent modification of frustrated magnetic interactions is consistent with our observation of an intimate correlation between the pressure-induced decrease in T_m and the associated increase in spin-disorder scattering [see Figs. 3(a) and 3(b)]. It would also imply the existence of a second QCP in YbAgGe around 16 GPa. In this context it is interesting to refer to very recent reports on the possibility of the occurrence of two quantum-critical points in a nonmagnetic Yb-based compound, Yb₂Pd₂Sn, located in the proximity to a magnetic QCP under high pressure.^{29,30} We feel that our results would stimulate further studies of the interplay between geometrical frustrations and long-range magnetic order in related strongly correlated electron systems.

In summary, we have investigated the magnetic and structural properties of the frustrated heavy-fermion antiferromagnet YbAgGe under high pressure up to about 20 GPa using electrical resistivity and x-ray diffraction, respectively. The high-pressure results showed that with increasing pressure, the magnetic ordering temperature (T_m) first rapidly increases, passes through a maximum ($T_m=5.4$ K at 6 GPa), and then drops toward zero for p above 16 GPa, while the lattice structure of YbAgGe remains unchanged up to 20 GPa. It is suggested that the anomalous pressure dependence of T_m is determined by a complex interplay of Kondo effect, RKKY and the geometrical frustration of the magnetic interactions of the Yb moments, and the relative strength of these low energy scales with increasing pressure. The sharp increase in T_m is supposed to be due to the suppression of the Kondo effect and spin fluctuations, which in turn leads to a gradual recovery of the Yb local magnetic moments. On the

other hand, we have shown that observed rapid decrease in T_m for $p > 6.8$ GPa and the disappearance of magnetic ordering for $p = 16.5$ GPa can be explained by a pressure-induced modification of the frustrated magnetic interactions in the Kagomé-type lattice structure of the YbAgGe. In this scenario, we have discussed the possible occurrence of a pressure-induced second quantum-critical point in YbAgGe

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