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Quantum Bicriticality in the Heavy-Fermion Metamagnet YbAgGe

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Bicritical points, at which two distinct symmetry-broken phases become simultaneously unstable, are typical for spin-flop metamagnetism. Interestingly, the heavy-fermion compound YbAgGe also possesses such a bicritical point (BCP) with a low temperature $T_{\rm BCP} \approx 0.3$ K at a magnetic field of $\mu_0 H_{\rm BCP} \approx 4.5$ T. In its vicinity, YbAgGe exhibits anomalous behavior that we attribute to the influence of a quantum bicritical point that is close in parameter space yet can be reached by tuning $T_{\rm BCP}$ further to zero. Using high-resolution measurements of the magnetocaloric effect, we demonstrate that the magnetic Grüneisen parameter Γ_H indeed both changes sign and diverges as required for quantum criticality. Moreover, Γ_H displays a characteristic scaling behavior but only on the low-field side $H \lesssim H_{\rm BCP}$, indicating a pronounced asymmetry with respect to the critical field. We speculate that the small value of $T_{\rm BCP}$ is related to the geometric frustration of the Kondo lattice of YbAgGe.

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Exotic quantum states of matter may arise in magnetic systems when long-range ordering of magnetic moments is suppressed by either competing interactions or geometrical frustration. A "strange metallic state," a so-called non-Fermi liquid, is found in magnetic metals close to a quantum critical point with competing Ruderman-Kittel-Kasuya-Yosida and Kondo interactions [1]. Local-moment systems with geometrical frustration, on the other hand, remain disordered down to zero temperature and form a strongly correlated paramagnet, i.e., a quantum spin liquid [2]. Usually, these two intriguing quantum states of matter are experimentally studied in different material classes: the former in heavy fermions and the latter in insulating spin systems. The current interest of research on heavy fermions, however, increasingly focuses on the interplay of these two phenomena [3,4], bridging two distinct fields of research. It has been proposed that frustrating the magnetic interaction in Kondo-lattice systems gives rise to a rich phase diagram that, as an exciting possibility, also contains unconventional, metallic spin-liquid phases [5,6].

For the exploration of magnetic frustration, heavy-fermion systems with a geometrically frustrated crystal lattice are of particular interest. The pyrochlore Kondo-lattice compound $Pr_2Ir_2O_7$, for example, indeed does not order magnetically down to lowest temperatures but exhibits anomalous thermodynamics [7] and, interestingly, a spontaneous Hall effect [8], which have been attributed to the formation of a metallic chiral spin liquid.

In this work, we investigate the heavy-fermion compound YbAgGe [9–12] that possesses a hexagonal ZrNiAl-type crystal structure, forming a two-dimensional distorted kagome lattice [13], which promotes frustration effects. At high temperatures, the magnetic susceptibility shows Curie-Weiss behavior with a Weiss temperature $\Theta_{ab} = -15$ K for a small magnetic field within the ab plane [9]. At low temperatures $T \lesssim 4$ K, antiferromagnetic correlations develop, reflected in a maximum in the susceptibility χ_{ab}. Finally, a sharp first-order transition into a magnetically ordered phase occurs at $T_N = 0.65$ K [9,10,12,14,15]. The large factor $f = |\Theta_{ab}|/T_N = 23$ indicates frustrated interaction between magnetic moments. This is corroborated by the structure factor of the damped spin fluctuations observed at low temperatures, which consists of sheets in reciprocal space corresponding to quasi-one-dimensional behavior along the c axis [16].

As a function of magnetic field applied within the ab plane, a complex phase diagram is found with various regions that were labeled by letters a to f in Ref. [14]. The regions a to d are interpreted as symmetry-broken phases because their boundaries have been located by anomalies in thermodynamics and transport [14,15,17], although some of them are only weak. The antiferromagnetic ordering wave vectors of the a, b, and c phases were identified by neutron scattering [16,18,19], whereas the order parameter of the d phase remains elusive. It is, however, the latter d phase that exhibits intriguing properties; see Fig. 1. The linear-T resistivity [20], persisting in the whole field interval of the d phase, indicates that it is a rare example of a genuine non-Fermi liquid phase. Furthermore, its American-muffin-shaped phase boundary implies a higher entropy than the one of the adjoining phases similar to the nematic phase in the metamagnet $Sr_3Ru_2O_7$ [21,22]. The anomalous behavior is most pronounced close to its lower boundary field at $\mu_0 H_{cd} = 4.8$ T, which was suggested to be caused by field-induced quantum criticality [10]. Recently, this was confirmed by the observation of a sign change in the thermal expansion and an incipient divergence of the Grüneisen parameter

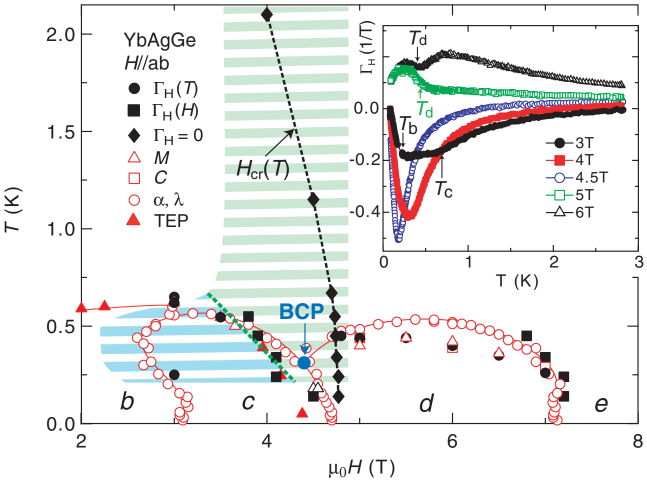

FIG. 1 (color online). H-T phase diagram of YbAgGe for fields within the ab plane focusing on the region with the BCP. Black and red symbols are obtained in this and previous studies [14,15,17], respectively. The solid red lines are guides for the eye. The dotted black line $H_{\mathrm{cr}}(T)$ shows maxima in entropy identified by the location of zeros of Γ_H (diamond). Solid black symbols denote anomalies in T sweeps (filled circle) and H sweeps (filled square) of Γ_H. Red symbols are transitions observed in magnetization (M, open triangle), specific heat (C, open square), thermal expansion coefficient and magnetostriction (α and λ, respectively, open circle) and thermoelectric power (TEP, filled triangle). Within the shaded area, Γ_H exhibits scaling behavior (see Fig. 4) with a crossover indicated by the dotted green line. The inset shows the magnetic Grüneisen ratio Γ_H as a function of temperature for fields applied parallel to the ab plane; arrows indicate phase transitions.

[14]. Furthermore, at this critical field, an anomalous reduction of the Lorenz ratio was observed, suggesting a violation of the Wiedemann-Franz law [23].

In Ref. [14], the origin of quantum criticality was attributed to a quantum critical endpoint (QCEP), i.e., an isolated endpoint of a line of first-order metamagnetic quantum phase transitions [24–26]. However, this scenario does not account for the presence of both symmetry-broken phases c and d close to the critical field H_{cd}. Similarly, a standard quantum critical point (QCP), whose phase diagram only involves a single symmetry-broken phase, can also be excluded. Interestingly, closer inspection of the phase diagram reveals that the phase boundaries of the c and d phases merge at a bicritical point (BCP) located at a temperature $T_{\mathrm{BCP}} \approx 0.3$ K and field $\mu_0 H_{\mathrm{BCP}} \approx 4.5$ T; see Fig. 1. Below T_{BCP}, a direct first-order transition between the two phases emerges, signaled by the appearance of hysteresis in magnetostriction [14]. As the hysteresis becomes very weak close to the BCP and the slope of the d-phase boundary is relatively steep so that its signatures are hard to follow in the thermal expansion [14], we estimated the location of the BCP with an uncertainty in T_{BCP} and $\mu_0 H_{\mathrm{BCP}}$ of about ± 0.1 K and ± 0.1 T, respectively.

Bicriticality typically arises in local-moment antiferromagnets with weak magnetic anisotropies resulting in spin-flop metamagnetism [27], as illustrated in Fig. 2(a). We envision that T_{BCP} can be tuned to zero by increasing the strength of frustration, giving rise to a quantum bicritical point (QBCP); see Fig. 2(b). Further increase of the frustration strength would naturally cause a separation of the two ordered phases, leading to a field-induced quantum spin-liquid state existing in a finite interval of magnetic field; see Fig. 2(c). We propose that in YbAgGe, such a QBCP is close in parameter space and at the origin of the pronounced anomalies observed near H_{cd}; see Fig. 2(d).

In order to further investigate the field-induced quantum bicriticality, we measured the magnetocaloric effect $(dT/dH)_S = T\Gamma_H$. The quantity Γ_H is a magnetic analogue of the Grüneisen parameter that necessarily diverges at a QCP [28,29]. Compared to the latter that was already determined for YbAgGe in Ref. [14], Γ_H has the important advantage that it does not necessitate additional assumptions about the magnetoelastic coupling, and it directly provides information about the entropy distribution within the H-T phase diagram via the relation $(\partial S/\partial H)_T = -\Gamma_H/C_H$. In particular, maxima in entropy are reflected in sign changes of Γ_H [29]. For the prototypical field-induced QCP material $YbRh_2Si_2$, a divergence of Γ_H at its critical field was reported in Ref. [30].

Single crystals were grown from high temperature ternary solutions rich in Ag and Ge [9]. The magnetocaloric effect was measured with very high resolution in a dilution

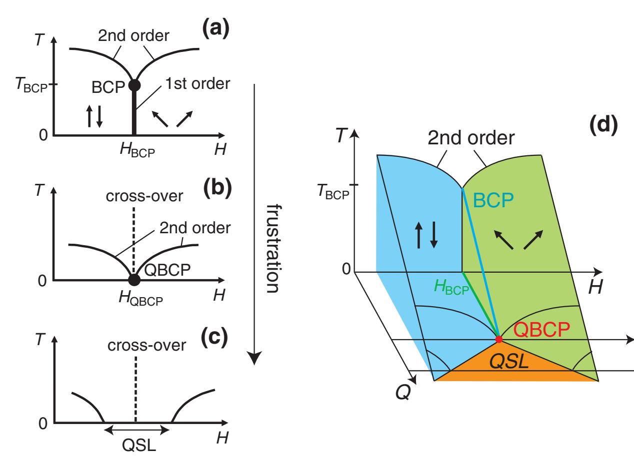

FIG. 2 (color online). Schematic phase diagrams with spin-flop bicriticality at different frustration strengths Q. (a) Bicritical point at a finite temperature $T_{\mathrm{BCP}} > 0$. (b) Frustration suppresses the transition temperatures and thus T_{BCP}, leading to a QBCP; the dotted line represents a crossover line separating two paramagnetic states with different short-range orders of frustrated moments. (c) Suppressing the transition temperature further results in a field-induced quantum spin liquid (QSL) present in a finite window of magnetic field. (d) Schematic T-H-Q phase diagram. Field-induced phase transition at H_{BCP} is of first order. QSL phase for strong frustration Q is indicated by orange color.

refrigerator with a superconducting magnet equipped with an additional modulation coil by utilizing the alternating field technique [31].

Temperature scans of Γ_H of YbAgGe are shown in the inset of Fig. 1 at different magnetic fields $3\ \mathrm{T} \leq \mu_0 H \leq 6\ \mathrm{T}$. The arrows indicate the anomalies associated with the phase transitions into the c and d phases. The most noticeable feature, however, is the spreading of the set of Γ_H curves as the temperature is lowered down to the transition temperatures with a sign change around H_{cd}. Such a behavior originates from an accumulation of entropy close to the critical magnetic field and is characteristic for metamagnetic quantum criticality [24–26]. At the low-field side $H < H_{cd}$ for $\mu_0 H = 4$ and 4.5 T, Γ_H is strongly temperature dependent and develops a pronounced peak whose height and width increases and decreases, respectively, as criticality is approached. The peak position for $\mu_0 H = 4.5$ T is located at 0.18 K well below the transition temperature, indicating that the quantum critical fluctuations are hardly quenched upon entering the ordered phase. In contrast, for $H > H_{cd}$, such a peak is absent as $\Gamma_H(T)$ is substantially suppressed upon entering the d phase, e.g., at 6 T.

Magnetic field sweeps of Γ_H for temperatures $T < 0.7$ K are shown in Fig. 3. Whereas the signatures at the transitions b-c and d-e are minor, the transition between c and d phases is clearly revealed. In the field sweep, the sign change of Γ_H close to the critical field H_{cd} is particularly evident. A sign change of Γ_H coincides with a maximum in entropy $S(H)$ as illustrated in the inset of Fig. 3. Here, the entropy was obtained by integrating $(\partial S/\partial H)_T = -\Gamma_H/C_H$ using the specific heat data of Ref. [15]. The position of sign changes $\Gamma_H = 0$, obtained from the T and

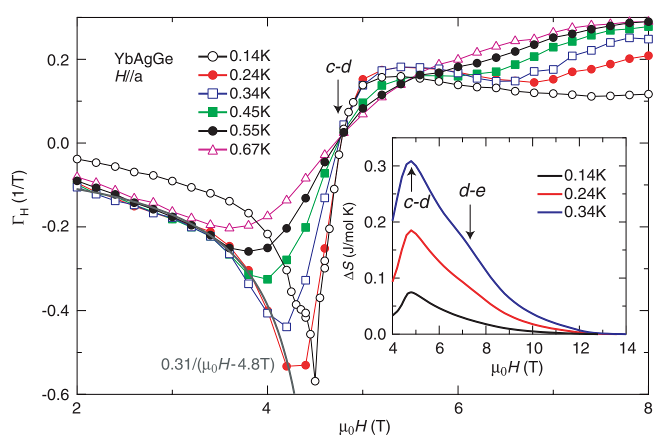

FIG. 3 (color online). Γ_H as a function of magnetic field applied parallel to the ab plane for low temperatures $T < 0.7$ K. The solid gray line is the power law $0.31/(\mu_0 H\text{-}4.8\ \mathrm{T})$. The inset shows a change of entropy calculated from Γ_H with the help of the reported specific heat [15] (shifted vertically such that $\Delta S = 0$ for $\mu_0 H = 13$ T). Arrows indicate the positions of phase transitions at $T = 0$.

H sweeps, defines the entropy ridge $H_{cr}(T)$ that is shown by the dotted black line in Fig. 1. In the low temperature limit, it extrapolates to the critical field $H_{cr}(T) \to H_{cd}$ for $T \to 0$. Its temperature dependence $H_{cr}(T)$ is rather weak, as it starts with an infinite slope. Nevertheless, it is considerably stronger (20% reduction at 2 K from its zero temperature limit) as compared to other itinerant quantum critical metamagnets like $CeRu_2Si_2$ ($\sim 0.5\%$ increase at 1.5 K) [25] and $Sr_3Ru_2O_7$ (no change up to 3 K) [21].

As already anticipated from the T sweeps, the field dependence of Γ_H around the critical field becomes highly asymmetric at low temperatures with a rounded shoulder for $H > H_{cd}$ but a pronounced negative peak on the low-field side. Strikingly, for temperatures in the range 0.24 up to 0.64 K, $\Gamma_H(H)$ first nicely traces a common curve $\Gamma_H \approx -G_H/(H - H_{cd})$ as expected for quantum criticality [28] with a fitted prefactor of $G_H \approx -0.31$. Closer to the critical field, it crosses over towards positive values with a sign change at the critical field that persists down to lowest temperatures. Finally, at a temperature $T = 0.14$ K, well below the bicritical point, Γ_H still exhibits a sharp decrease towards the critical field but deviates from the common scaling curve.

In the case of a metamagnetic QCEP, an Ising symmetry emerges, resulting in symmetric behavior between low and high fields [25]. Such an approximate symmetry is observed in $Sr_3Ru_2O_7$ [21], $CeRu_2Si_2$ [25], and $Ca_{2-x}Sr_xRuO_4$ [32]. The QBCP also terminates a line of first-order transitions that, in contrast to the QCEP, separates, however, two distinct symmetry-broken phases. As a consequence, quantum bicritical behavior is generically expected to be asymmetric with respect to the critical field, which apparently applies to YbAgGe. Such an asymmetry might be induced, in particular, by distinct dynamical exponents z for critical fluctuations associated with the two adjacent phases, so that the QBCP is generally characterized by multiple dynamics [33,34].

In order to investigate the properties of the QBCP in YbAgGe quantitatively, we proceed with analyzing the scaling of Γ_H observed experimentally. As the critical signatures for high fields are rather weak, we concentrate in the following on the scaling that is apparent on the low-field side $H < H_{cd}$. Similarly, as suggested for the QCEP [26], we consider two scaling parameters given by temperature T and $h = H - H_{cr}(T)$, i.e., the distance in field to the location of entropy maxima $H_{cr}(T)$. The weak T dependence of $H_{cr}(T)$ is irrelevant in the limit $T \to 0$, but the extracted scaling sensitively depends on it. Using the temperature-dependent scaling field h, it is possible to reveal scaling behavior in a larger temperature regime otherwise hidden by the temperature drift of the entropy ridge $H_{cr}(T)$.

For data on the low-field side $H < H_{cd}$ and $T > 0.2$ K within the shaded regime of Fig. 1, we find that Γ_H obeys $T/|h|^{1.1}$ scaling behavior; see Fig. 4. The linear scaling plot

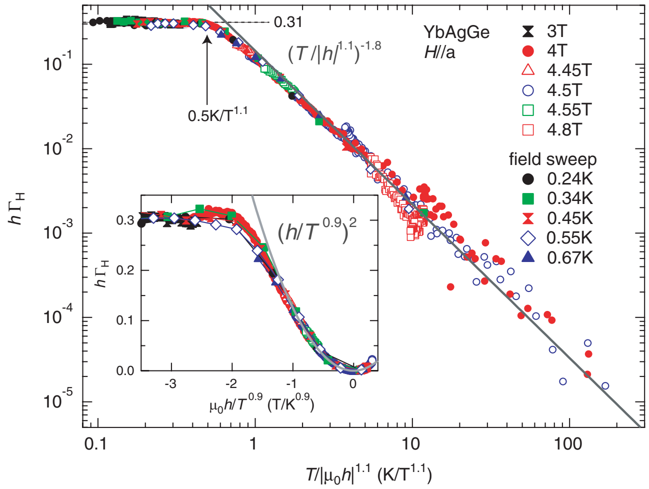

FIG. 4 (color online). Scaling plot of $h\Gamma_H$ vs $T/|h|^{1.1}$ for data only within the shaded regime of Fig. 1 with $h = H - H_{cr}(T)$, where $H_{cr}(T)$ is defined by the zeros of Γ_H (dotted line in Fig. 1). An arrow indicates the crossover at 0.5 K/T$^{1.1}$ of the scaling function also shown by the dotted green line in Fig. 1. The solid gray line represents the asymptotics $\Gamma_H \sim h/T^{1.8}$. The inset shows the scaling plot on a linear scale but as a function of $h/T^{0.9}$.

shown in the inset of Fig. 4 displays a high quality of collapse also at high values of $h\Gamma_H$. The scaling can be described by a function of the form $\Gamma_H = \frac{1}{h}\mathcal{G}(h/T^{1/(\nu z)})$ with an exponent $\nu z = 1.1$. The excellent collapse of data with the absolute temperature T as a scaling field confirms in particular that the anomalous behavior around H_{cd} is indeed caused by a *quantum* (bi-)critical point and not by the classical counterpart at $T_{BCP} \approx 0.3$ K. For classical bicriticality instead data collapse in terms of the differences $T - T_{BCP}$ and $H - H_{BCP}$ would be expected.

Two scaling regimes separated by a crossover at $T/|\mu_0 h|^{1.1} \approx 0.5$ K/T$^{1.1}$ can be distinguished. For low temperatures and large negative h, $\Gamma_H h$ approaches a constant with the value $-G_H = 0.31$. For high temperatures and small h, on the other hand, it is expected that Γ_H vanishes analytically, i.e., linearly with h because the line $H_{cr}(T)$ is only a crossover where thermodynamics remains smooth [35]. Analyticity thus requires the function $\mathcal{G}$ to behave for small arguments as $\mathcal{G}(x) \propto x^2$. This determines the asymptotics $\Gamma_H \sim h/T^{2/(\nu z)} \approx h/T^{1.8}$, which is indeed observed at high temperatures, as indicated by the solid line in Fig. 4.

Interestingly, in the mixed-valence compound β-$YbAlB_4$, a T/h scaling with a similar exponent ($\nu z = 1$) and a divergence $\Gamma_H \sim h/T^2$ was observed but with $H_{cr} = 0$ [36]. Effective one-dimensional degrees of freedom have been invoked there for its explanation [37]. Quasi-one-dimensional fluctuations have already been observed in YbAgGe but in zero field probably promoted by the geometrical frustration [16]. This low dimensionality of fluctuations might also survive in finite field and drive the critical behavior at H_{cd}. A hint in this direction is provided by the strong temperature dependence of the magnetization $(\partial M/\partial T)_H$ [15] and the thermal expansion α [14] that both behave as $\sim h/T^{1.7}$ close to the critical field [38]. From a quantum critical scaling point of view, such a strong divergence as a function of T implies a low spatial dimensionality d [28].

A neutron scattering study in the bicritical regime would be useful not only to determine the dimensionality but also the dynamics of the critical fluctuations. This might also shed light on the pronounced asymmetry of the quantum bicritical behavior with respect to the critical field. Moreover, in order to construct a basic theory of the QBCP and to test its spin-flop character, the experimental identification of the order parameter of the d phase is mandatory. Further experimental and theoretical work will be required to elucidate the origin of the QBCP and the observed scaling exponents.

To summarize, we propose that YbAgGe is situated close to a QBCP that controls thermodynamics in its vicinity. We verified that the magnetic Grüneisen parameter Γ_H exhibits the corresponding quantum critical signatures and identified a characteristic scaling behavior.

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