High temperature antiferromagnetism in Yb based heavy fermion systems proximate to a Kondo insulator

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Given the parallelism between the physical properties of Ce and Yb based magnets and heavy fermions due to the electron-hole symmetry, it has been rather odd that the transition temperature of the Yb based compounds is normally very small, as low as ~ 1 K or even lower, whereas Ce counterparts may often have the transition temperature well exceeding 10 K. Here, we report our experimental discovery of the transition temperature reaching 20 K for the first time in a Yb based compound at ambient pressure. The Mn substitution at the Al site in an intermediate valence state of α-YbAlB₄ not only induces antiferromagnetic transition at a record high temperature of 20 K but also transforms the heavy fermion liquid state in α-YbAlB₄ into a highly resistive metallic state proximate to a Kondo insulator.

Correlated electron systems have provided a number of non-trivial phenomena including quantum criticality, unconventional superconductivity and exotic magnetic/electronic orders. Specifically, 4f electron systems have provided prototypical materials to study quantum criticality that appears as a result of the competition between the RKKY interaction and the Kondo effect [1–11]. 4f electron systems also provide ideal platforms to investigate the effects of correlation in the presence of the strong spin-orbit coupling. The discovery of novel topological phases in correlated electron systems such as topological insulators and Weyl semimetals progressively attracted attention to even more strongly correlated system [12–17], including 4f electron systems with, for example, the so-called topological Kondo insulators [18–21].

Among all lanthanide ions, most studies have focused on Ce and Yb based compounds as they are at the one electron and the one hole limits of the 4f shell respectively. This electron-hole symmetry implies a parallelism between the physical properties of Ce and Yb based magnets and heavy fermions. However, the transition temperature of the Yb based compounds is normally very small, as low as ~ 1 K or even lower [3, 5, 22–31], whereas Ce counterparts may have the transition temperature often well exceeding 10 K [32–36].

Among a number of Yb-based heavy fermion systems, β-YbAlB₄ is particularly interesting as it is the first example of a heavy fermion superconductor and unconventional quantum criticality without tuning, namely, at ambient pressure and at zero field [8, 37–39]. In contrast, its isomorphic compound α-YbAlB₄ has a Fermi liquid ground state while it exhibits almost the same Kondo lattice like behavior at higher temperatures [40]. Recent study revealed that a small amount of Fe doping of 1.4% induces nearly the same type of quantum criticality as found in β-YbAlB₄ at ambient pressure [41]. The clear anomaly in the low temperature valence indicates quantum valence transition as the most likely origin [41–43].

Another particularly striking feature found in these compounds is the unusually high transition temperature of their magnetism. For example, the transition temperature exceeding 30 K was observed for β-YbAlB₄ under high pressure, which is a record high transition temperature for Yb based compounds to date [44, 45]. In addition, for both α and β phases of YbAlB₄, a sufficient Fe doping stabilizes an antiferromagnetic ordering whose transition temperature becomes as high as 10 K at ambient pressure [41, 46, 47]. These fascinating observations of the unusually high transition temperature as well as the unconventional quantum criticality in these systems indicate a novel mechanism behind the phenomena. A significant question would be whether the transition temperature may become even higher than 10 K by another type of chemical doping to these systems. If so, such
FIG. 1. (color online). Phase diagram as a function of temperature $T$ (vertical axis) and Mn concentration $x$ (horizontal axis) for $\alpha$-YbAl$_{1-x}$Mn$_x$B$_4$. The contour plot of the $ab$-plane resistivity $\rho$ is also provided in the same region of $T$ and $x$. Each point indicates the Néel temperature $T_N$ determined by the anomaly found in the temperature dependence of the magnetization (squares) and specific heat (circles). The broken line schematically refers to an phase boundary due to the antiferromagnetic transition. Inset indicates the unit cell crystal structure of $\alpha$-YbAl$_{1-x}$Mn$_x$B$_4$.

a study would not only find the highest ever magnetic transition temperature in the Yb-based compounds at ambient pressure, but it also allows further in-depth study to elucidate the electronic state change across the transition by spectroscopic method, which has been otherwise impossible for Yb compounds to date.

Here, we report the discovery of a high Néel temperature reaching 20 K, the highest temperature among the Yb-based intermetallic compounds at ambient pressure. As we show in Fig. 1, the Mn substitution at the Al site of 4\% is enough to induce an antiferromagnetic transition in $\alpha$-YbAlB$_4$. The transition temperature exceeds 20 K at the doping level of $\sim$ 30\% Mn. The high transition temperature cannot be explained by the RKKY interaction and thus points to an itinerant type magnetic order. On the other hand, the combination of the transport and the photoemission spectroscopy (PES) measurements reveals that the Mn doping induces a hybridization gap at the Fermi energy ($E_F$), causing a highly resistive nonmetallic behavior near $x$(Mn) = 0.5. Our study indicates that the extremely high transition temperature appears in a heavy fermion (HF) state proximate to a Kondo insulator phase.

Figures 2(a) and 2(b) display the temperature dependence of the $ab$-plane magnetic susceptibility and specific heat, respectively. With increasing Mn concentration $x$, the temperature dependence of the susceptibility exhibits a clear kink and a bifurcation between the zero field cooled (ZFC) and field cooled (FC) components of the magnetic susceptibility. While no magnetic anomaly is found at $x = 0.01$, the bifurcation is clearly seen at the concentration $x$ higher than 0.11, and a small anomaly appears already at $x = 0.07$. Correspondingly, the specific heat measurement finds a clear peak at almost the same temperature as the bifurcation temperature of the magnetic susceptibility curve. The anomaly observed in both the susceptibility and specific heat demonstrates that the antiferromagnetic transition is induced by the Mn substitution. Strikingly, the transition temperature ($T_N$) increases with increasing chemical substitution and reaches a temperature as high as 20 K at $x = 0.27$. On the other hand, the susceptibility measured for the non-magnetic analog $\alpha$-LuAl$_{0.56}$Mn$_{0.44}$B$_4$ has a much smaller value around $\chi \sim 2 \times 10^{-4}$ emu/mol and its temperature dependence does not exhibit any anomaly. All these results including a $\mu$SR measurement (given in the Supplemental Material [48]) indicate that Mn in $\alpha$-LuAl$_{0.56}$Mn$_{0.44}$B$_4$ should be nonmagnetic.

To our knowledge, the antiferromagnetic transition temperature reaching 20 K is the highest among the Yb-based magnets and HF compounds at ambient pressure. Interestingly, $T_N$ peaks at $x = 0.27$ and starts decreasing with further substitution as found in both magnetic susceptibility and specific heat results [Figs. 2(a) and 2(b)]. It should be also noted that the Mn substitution changes the magnetic anisotropy. Whereas the pure $\alpha$-YbAlB$_4$ has the Ising type moment along the $c$-axis and the $ab$-plane susceptibility is almost temperature independent below 300 K, the Mn substitution induces a more isotropic behavior with a strong temperature dependence in the $ab$-plane, as visible in the inverse susceptibility in the inset of Fig. 2(a).

To further understand the Mn substitution effects, we fitted both the $ab$-plane and the $c$-axis components of the susceptibility $\chi(T)$ using the Curie-Weiss (CW) law, $\chi = \frac{N_x\mu_B^2}{k_B(T - \Theta_W)}$, where $k_B$, $\Theta_W$, $N_x$, $\mu_B$ and $\mu_B$ refer the Boltzmann constant, the Weiss temperature, the Avogadro’s number, the effective moment and the Bohr magneton, respectively. The fitting was performed in the temperature range from 150 K to 250 K except for the $ab$-plane component of the pure system [Fig. 2(a) inset]. On the other hand, the magnetic part of the entropy $S_{mag}$ was estimated by $S_{mag} = \int C_{mag}dT$, assuming a linear increase of $C_{mag}$ between 0 and 2 K. Here, $C_{mag}$ indicates the magnetic part of the specific heat estimated as noted in the caption of Fig. 2(b). Interestingly, the entropy in the paramagnetic state, e.g. at 40 K, increases with increasing Mn substitution and reaches a value of about $R \ln 2$ for $x \geq 0.27$ [Fig. 2(b) inset]. This indicates that the ground state remains a doublet, but the associated Kondo temperature $T_K$ decreases significantly with increasing Mn substitution as we discuss in the following.

Figure 2(c) shows the $x$(Mn) dependence of the Kondo temperature $T_K$, of the Weiss temperature $\Theta_W$ and of
the effective moment $P_{\text{eff}}$. $T_K$ was estimated from the temperature dependence of the entropy [Fig. 2(b) inset] using the relation $S_{\text{mag}} (T = T_K/2) = (R/2) \ln 2$. Both the Kondo temperature $T_K$ and the Weiss temperature $\Theta_W (c)$, extracted from the $c$-axis component of the susceptibility, nearly collapse with increasing $x$(Mn). In its $ab$-plane component, the susceptibility of the pure system ($x = 0$) exhibits a peak at about 200–250 K due to the significant valence fluctuations in this system [40, 43, 49]. Therefore, we extended our susceptibility measurements up to 700 K to find a CW behavior above 300 K [Fig. 2(a) inset]. While both $T_K$ and $\Theta_W (c)$ are found to be of about $\sim 130$ K, $\Theta_W (ab)$ reaches about 350 K in the pure system from the fitting of the $ab$-plane component of the susceptibility in between 600 K and 700 K. With increasing $x$(Mn), we find a systematic decrease in all $\Theta_W (ab)$, $\Theta_W (c)$ and $T_K$. This indicates the valence fluctuation regime in $\alpha$-YbAlB$_4$ is suppressed by the Mn substitution. In fact, the local moment $P_{\text{eff}}$ gradually increases to reach $4.5 \sim 4.7 \mu_B$ at $x > 0.27$, a value equivalent to the one of the isotropic Yb$^{3+}$ ion: $4.54 \mu_B$. Thus the Mn substitution leads the Yb ions to a trivalent configuration.

Generally, in the Kondo regime, the magnetic couplings are dominated by the RKKY interaction. Theoretically, the magnetic transition temperature due to the RKKY interaction is known to be proportional to the de Gennes factor [50] (see the Supplemental Material [48]). However, $T_N$ of $\alpha$-YbAl$_{0.73}$Mn$_{0.27}$B$_4$ is strikingly one order of magnitude higher than those with other RAIB$_4$ systems having the YCrB$_4$ structure [51, 52]. Namely, the magnetism in $\alpha$-YbAl$_{1-x}$Mn$_x$B$_4$ is not a localized RKKY magnetism, but most likely an itinerant type.

An important effect to understand the origin of the itinerant magnetism is the increase in the hybridization strength with the Mn substitution. The substitution decreases the volume of the unit cell [48], and thus applies chemical pressure to the system. The valence change discussed above from the larger Yb$^{2+}$ to smaller Yb$^{3+}$ ions also causes a similar trend. Therefore, the bond lengths between Yb and B become shorter, and thus the hybridization strength should increase with the Mn substitution [53, 54]. Further evidence for the itinerant magnetism will be discussed below using the results obtained in the resistivity and PES measurements.

Recent structure analysis using powder X-ray diffraction under high pressure on $\beta$-YbAlB$_4$ indicates that the B rings sandwiching Yb ion (Fig. 1 inset) lowers its symmetry from nearly 7 fold by application of pressure [55]. One theoretical approach pointed out that this 7 fold symmetry stabilizes the Yb ground state to $|J_z = \pm 5/2 >$. This orbital character is shown to imply an anisotropic hybridization as well as the Ising behavior of the magnetic moment along the $c$-axis at ambient pressure [56, 57]. These results suggest that the deformation of the local symmetry under pressure suppresses the Ising-like behavior and induces the 30 K transition under 8 GPa in $\beta$-YbAlB$_4$.

This may be also the case in $\alpha$-YbAlB$_4$ and the magnetism induced by the Mn substitution. Nearly the same temperature dependence of the susceptibility has been seen for both phases of YbAlB$_4$ at ambient pressure. Thus, the crystal electric field (CEF) scheme must be nearly the same for both phases [40]. Indeed, a recent measurement of X-ray magnetic circular dichroism indicates the ground state of $\alpha$-YbAlB$_4$ to be the $|J_z = \pm 5/2 >$ state [49]. On the other hand, the systematic change in anisotropy of the susceptibility as visible in
the inset of Fig. 2(a) and in Fig. 2(c) demonstrates that the CEF ground state changes with the Mn substitution, while keeping a doublet configuration as the magnetic entropy at 40 K remains around $R \ln 2$. Thus, most likely the ground doublet departs from the $|J_z = \pm 5/2\rangle$ state due to the local symmetry deformation around the Yb site, itself induced by the Mn substitution. This change of ground-state plays an important role in the high temperature magnetism, as discussed below.

Another effect of the chemical substitution is the carrier doping. Such carrier doping may well change the transport properties, and therefore, we performed the longitudinal resistivity measurements [Fig. 2(d)]. The magnitude as well as the temperature dependence of the resistivity dramatically change with the Mn substitution. The good metallic behavior observed in the pure system is already lost at $x = 0.11$ and the resistivity at both room temperature and 2 K increases nearly linearly with $x$. The resistivity at 2 K for $x = 0.57$ is $\sim 60$ times larger than the one for $x = 0$. Moreover, the temperature dependence systematically changes into a more incoherent semimetallic or non-metallic behavior with Mn doping. These indicate that the carrier concentration must decrease with increasing Mn concentration. We also point out that the high temperature resistivity for $x \sim 0.5$ can be roughly fitted with the activation law, namely, $\rho = \rho_0 \exp(\Delta/T)$ with an activation energy of about $\Delta \sim 50$ K. The inset of Fig. 2(d) exemplifies such a fit with the magnetic part of the resistivity for $x = 0.48$ in which the activation law is observed at high temperature from 200 K to above 300 K. This suggests that the highly resistive transport induced by Mn substitution results from the system approaching an insulating state. The deviation from the activation law at low temperature indicates the presence of impurity bands which cause the saturation of the resistivity on cooling. In addition, a clear anomaly at $T_N$ can be found for the high $T_N$ samples with $x = 0.11$ and 0.27. Ce(Ru$_{0.85}$Rh$_{0.15}$)$_2$Si$_2$ exhibits a quite similar behavior at a spin-density-wave (SDW) transition [58]. This suggests the SDW transition as the origin of the anomaly seen between 0.04 $\leq x \leq 0.27$.

To investigate the origin of the insulating behavior, the density of states (DOS) near $E_F$ was examined using the high-resolution and bulk-sensitive laser PES [59, 60]. In order to evaluate the DOS below and slightly above $E_F$, the PES spectra were divided by spectra measured on a gold reference at each measured temperature. We should stress that this method is accurate only up to $5k_B T$ above $E_F$ [64], as indicated by the shaded energy range of Fig. 3(a). The derived results, referred as experimental DOS (ExDOS), scarcely depends on the temperature for $x = 0$ as shown by the inset of Fig. 3(a). On the other hand, for $x = 0.39$, ExDOS near $E_F$ is mostly flat at $T = 75$ K and gets suppressed as the temperature decreases. For quantitative discussion, ExDOS at $E_F$ is plotted for each measured doping in Fig. 3(b) as a function of temperature. For $x = 0$, ExDOS at $E_F$ exceeds 1, indicating a metallic state, in agreement with previous angle resolved PES measurements [61]. In contrast, ExDOS at $E_F$ for the doped samples ($x > 0$) falls below 1 and decreases when the temperature is lowered. The temperature-dependent depression of ExDOS is observed for all of the doped samples and starts far above the magnetic transition temperature $T_N$, although the spectral change appears larger around $T \sim 15$ K. These results are consistent with the resistivity measurements and indicate the formation of a pseudo-gap, or a $c$-$f$ hybridization gap, for $x > 0$. One may notice that, for $x = 0.39$, ExDOS across $E_F$ is highly asymmetric and finite ($>0$) at $E_F$ even at $T = 5$ K. A theoretical calculation based on the periodic Anderson model with $c$-$f$ hybridized bands [62, 63] can reproduce such DOS near $E_F$ at $T = 5$ K as also plotted in Fig. 3(a) [48]. In addition, DOS at $E_F$ reaches 0 in the calculation for $T = 0$ K using the same parameters set for $T = 5$ K, without any experimental broadening factor. This theoretical modeling suggests the gap formation observed with Mn doping to emerge from a Kondo-like hybridization. Since the gap opens over $E_F$, it naturally relates to the systematic increase of resistivity and pictures a crossover from an intermediate valence metal to a Kondo insulator as $x$ is substituted with Mn. Such Kondo-like hybridization further implies that the Yb 4$f$-electron bands around $E_F$ directly contribute to the transport and thus to the itinerant magnetism of $\alpha$-YbAl$_{1-x}$Mn$_x$B$_4$. The spectral shape and gap formation observed in ExDOS are further discussed in the Supplemental Material [48].

![FIG. 3. (color online). (a) Experimental density of state (ExDOS) near $E_F$ for $\alpha$-YbAl$_{1-x}$Mn$_x$B$_4$ ($x = 0.39$) derived from the PES measurements. The dotted lines indicate the calculation based on the periodic Anderson model [48]. The uncertain energy region on ExDOS over $\sim 5k_B T$ above $E_F$ [48, 64] is shaded (over $\sim 2.2$ meV at 5 K). Inset shows ExDOS for $x = 0.0$. ExDOS for other $x$ is displayed in the Supplemental Material [48]. (b) Temperature dependence of ExDOS at $E_F$ for various compositions.](image-url)
shows a high magnetic transition temperature which motivated intensive studies to reveal its origin [36, 70]. The similar highly resistive state, proximate to a Kondo insulator, found in $α$-YbAl$_{1-x}$Mn$_x$B$_4$ suggests that the same type of mechanism is at the origin of the high temperature magnetism.

Overall results indicate that the high temperature magnetism in this system is the itinerant SDW-type induced by chemical substitution, similar to the Rh substitution case in CeRu$_2$Si$_2$ [58]. The hybridization strength increases with the Mn substitution, while the suppression of the valence fluctuations in the Kondo regime – with an almost trivalent Yb configuration – leads to the decreases of the Kondo and Weiss temperatures. Theoretically, the strong hybridization may lead to a SDW-type order near the onset of a Kondo insulating phase [71]. The sharp coherent feature in PES seen only for the high $T_N$ samples between $x = 0.11$ and 0.39 [48] may result from the nesting across $E_F$ leading to a SDW-order.

The itinerant 4$f$-electrons in pure $α$-YbAlB$_4$ form quasi-two-dimensional bands. They do not contribute much to transport along the c-axis but do mainly in the $ab$-plane [40]. This suggests that the SDW-type antiferromagnetism is formed with the nesting vector in the $ab$-plane. The suppression of the magnetic anisotropy found in the susceptibility measurements by Mn doping indicates that the CEF, which initially implies the Ising-like ground state along the $c$-axis of the pure system, is modified by the distortion of the 7-fold local symmetry at the Yb site. As a result, the 4$f$ magnetic moment may develop a component in the $ab$-plane. As well known for the spin-flop transition in Cr [72], the SDW energy scale can be affected by the spin configuration and by its nesting vector. Therefore, the high $T_N$ induced by Mn substitution might also arise from the change in the CEF scheme. Further studies of the CEF scheme, both experimental (e.g. neutron scattering, NMR measurement) and theoretical, are necessary to clarify such possible mechanism.

To conclude, through the comprehensive measurements of $α$-YbAl$_{1-x}$Mn$_x$B$_4$, we found that the Mn substitution induces a high temperature antiferromagnetism, whose transition temperature reaches 20 K at $x = 0.27$. This is so far the highest magnetic transition temperature among the Yb based HF systems at ambient pressure. Our transport measurements show that the system exhibits a high resistivity with the Mn substitution. Thanks to PES, we further highlight the formation of a gap at the Fermi level, together with a coherent feature, consistent with the resistivity measurements. These observations evidence that the Mn substitution results in an increase of the Kondo-like hybridization, driving the system out of the mixed valence state of the pure $α$-YbAlB$_4$ to a more localized Kondo HF state. Additionally, the onset of the gap formation in the PES measurements appears at temperature well above the magnetic transition temperature; it lifts any ambiguity about the origin of the gap and underlines the crossover from a heavy Fermi liquid to a Kondo insulator with increasing Mn doping.

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