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Magnetic ordering in the pressure-stabilized high-temperature phase of $YbInCu_4$

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To elucidate the ground state of the pressure-stabilized high-temperature (HT) phase of $YbInCu_4$, we have carried out electrical resistivity ρ and ac-susceptibility χ_{ac} measurements at high pressures. For pressures above 2.49 GPa, the first-order valence transition is completely suppressed (below ~80 mK). Separately, above 2.39 GPa, a clear peak appears in χ_{ac} with a small kink in ρ at around $T_M = 2.4$ K. The χ_{ac} peak is easily diminished by applying low magnetic fields and disappears above ~500 Oe. The characteristic behavior of χ_{ac} at T_M can generally be ascribed to the onset of long-range ferromagnetic ordering and, therefore, the ground state of the pressure-stabilized HT phase is most probably a ferromagnetically ordered state. This result is compatible with the occurrence of weak ferromagnetism recently reported for the Y-substituted compound $Yb_{0.8}Y_{0.2}InCu_4$ under pressure of 1.2 GPa.

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The unstable $4f$ shell in intermetallic compounds containing Yb or Ce elements leads to a wide variety of physical properties, and recently the pressure-induced magnetically ordered and/or superconducting ground states have been the subject of great interest. Due to the hole-electron analogy, it has been anticipated that application of pressure in Yb-based compounds has an opposite effect compared to Ce based compounds, tending to induce a magnetically ordered state from a nonmagnetic state. However, the effect of pressure on Yb compounds has been less studied relative to the intensive studies on Ce compounds. This was mainly due to the rather high pressure required to tune the magnetic ordering: $P > 20$ GPa for YbCuAl;[1] $P > 8$ GPa for $YbCu_2Si_2$;[2] $P > 8$ GPa for Yb_2Ni_2Al;[3] and $P > 5$ GPa for $YbNi_2Ge_2$.[4]

A set of isostructural (C15b-type) ytterbium-based compounds $YbXCu_4$ ($X =$ Au, Cu, Ag, In, Cd, Tl, Mg) has a rich variety of ground states (localized spin, heavy-fermion, and fluctuating valence) that are strongly dependent on the species of X atoms.[5] Among the $YbXCu_4$ series, $YbInCu_4$ exhibits the temperature-induced first-order valence transition at $T_V \simeq 42$ K,[6,7] which is similar to the pressure-induced α-γ valence transition in Ce metal.[8] In the high-temperature (HT) phase of $YbInCu_4$, the magnetic susceptibility χ shows Curie Weiss-type spin paramagnetic behavior with an effective moment near the free Yb^{3+} ion value. In the low-temperature (LT) phase below the valence transition, the Yb valence is reduced to approximately 2.9 with a unit-cell volume expansion of 0.5 %.[7,9] The system transforms into the strongly enhanced Pauli paramagnetic state.[10] The Kondo temperature T_K for each of the HT and LT phases is estimated as ~25 and ~400 K, respectively.[5]

Both pressure and magnetic field can be used to induce the valence transition in $YbInCu_4$ from the nonmagnetic LT phase ($Yb^{2.9+}$) with larger cell volume to the magnetic HT phase (Yb^{3+}) with smaller one.[11] Uchida *et al.* performed resistivity measurements under pressure up to 3.3 GPa, and found that T_V is lowered to a temperature below 1.5 K at 2.5 GPa.[12] Yoshimura *et al.* observed at 5 K a sharp jump in the magnetization and negative volume magnetostriction of $\Delta V/V \sim -0.45\%$ at around ~30 T.[13]

Our interest is focused on the ground state realized in the magnetic HT phase after the valence transition is suppressed by pressure. Svechkarev *et al.* measured the magnetic susceptibility of $RInCu_4$ ($R =$ Gd, Er, and Yb) up to 0.2 GPa. Assuming a linear extrapolation of their data for $YbInCu_4$ to a higher-pressure region, they predicted that, with increasing pressure, the negative Weiss temperature at ambient pressure approaches the positive value which originates in the background interaction, so that ferromagnetic ordering appears.[14] For the Y substituted compound of $Yb_{0.8}Y_{0.2}InCu_4$, Mitsuda *et al.* recently reported that the collapse of the valence transition and a ferromagnetic ordering occur at 0.8 GPa almost simultaneously, and the weak ferromagnetic phase at 1.2 GPa is characterized by a low Curie temperature of 1.7 K.[15] To elucidate the ground state of the pressure-stabilized HT phase of pure $YbInCu_4$, we have carried out electrical resistivity ρ and ac susceptibility χ_{ac} measurements at high pressures up to 2.58 GPa and at low temperatures down to ~80 mK. Above 2.39 GPa, we found a clear peak in χ_{ac} at around $T_M = 2.4$ K with a small kink in ρ, which can be ascribed to the onset of long-range magnetic ordering. In the low temperature region, both at 2.49 and 2.28 GPa which are just above and below the critical pressure, ρ shows a monotonous decrease down to ~80 mK without any sign of superconductivity occurrence. Taken together, these data yield a new temperature-pressure phase diagram for $YbInCu_4$.

Single crystals of $YbInCu_4$ were grown using a flux technique as described in the literature.[16] To measure ρ and χ_{ac} at exactly the same pressure, both a single crystal with four fine

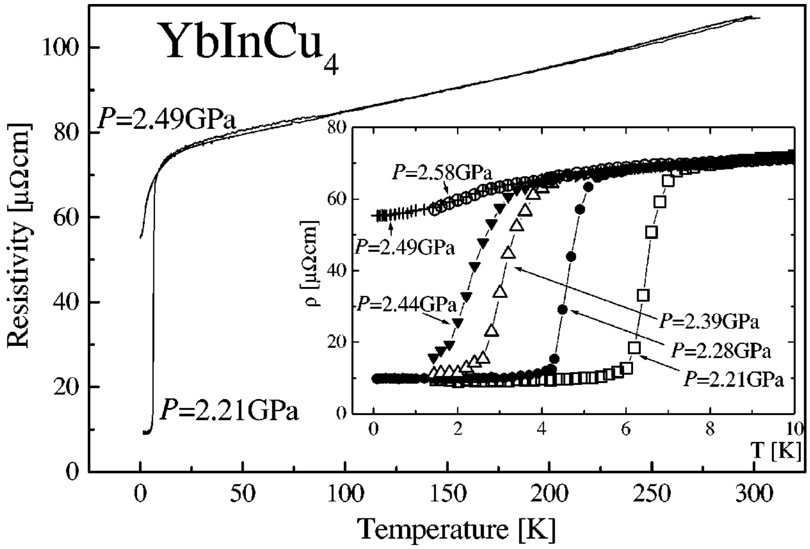

FIG. 1. Temperature dependence of the electrical resistivity of $\mathrm{YbInCu_4}$ under various pressures. The inset shows the data below 10 K.

wires of gold for the ρ measurement and a pick-up coil filled with many small crystals for χ_{ac} measurement were mounted inside a piston cylinder pressure cell constructed of nonmagnetic NiCrAl/BeCu. The pressure cell was filled with Daphne oil (7373) as a transmitting medium for hydrostatic pressure. For the experiment below 1 K, the pressure cell was assembled in a $^3\mathrm{He}/^4\mathrm{He}$ dilution refrigerator. The value of pressure was determined by monitoring the superconducting transition of a tin manometer. The electrical resistivity was measured by a four-probe ac resistance bridge (Linear Research, LR-700). The ac susceptibility was measured by the conventional method at a frequency of 132 Hz using primary and compensated pick-up coils mounted inside the pressure cell.

Figure 1 shows the temperature dependence of the electrical resistivity at pressures of 2.21, 2.28, 2.39, 2.44, 2.49, and 2.58 GPa. Above ~40 K, ρ exhibits almost T-linear behavior and hardly depends on the applied pressures. With decreasing temperature below ~30 K, ρ begins to deviate from the T-linear behavior. The sharp drop in ρ observed at lower temperatures for pressures between 2.21 and 2.44 GPa (inset of Fig. 1) originates from the valence transition from the HT to LT phases. The valence transition temperature $T_V \simeq 42$ K at ambient pressure is significantly reduced by applying pressure. (The dependence of T_V on pressure is shown in Fig. 4 as discussed later.) The value of $dT_V/dP \simeq -18.5$ K/GPa obtained for $P > 2.0$ GPa is comparable with the value reported previously.[12] At higher pressures above 2.49 GPa, ρ shows no sign of the valence transition down to ~80 mK, but a small kink at around $T = 2.4$ K instead. This new anomaly in ρ is obvious in T derivative of the resistivity $d\rho/dT$ as shown in Fig. 2(a) where we present a typical example for this pressure range. Later we will discuss the temperature dependence of ρ following T^2 as seen in Fig. 2(b).

Figures 3(a)–3(c) show the temperature dependence of χ_{ac} and ρ measured at $P = 2.21$, 2.39, and 2.49 GPa, respectively. At 2.21 GPa, χ_{ac} shows a small decrease below $T \sim 7$ K where ρ exhibits a simultaneous step-decrease. The small decrease in χ_{ac} can be associated with the bulk valence transition from the magnetic HT phase to the nonmagnetic LT phase. On the other hand, above 2.49 GPa, χ_{ac} exhibits a clear peak at around $T = 2.4$ K where ρ has the small kink

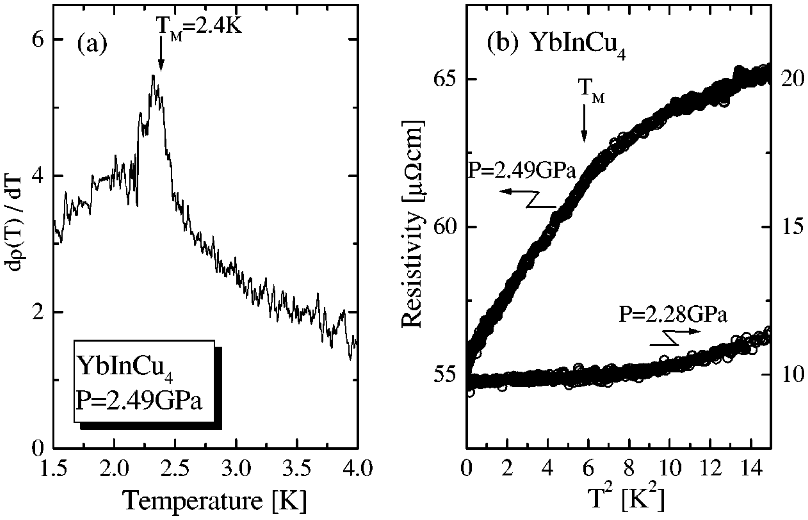

FIG. 2. (a) Dependence of the tempeature derivative of the electrical resistivity $d\rho/dT$ on the temperature for $\mathrm{YbInCu_4}$ at 2.49 GPa. (b) The electrical resistivity versus T^2 plot for 2.28 and 2.49 GPa.

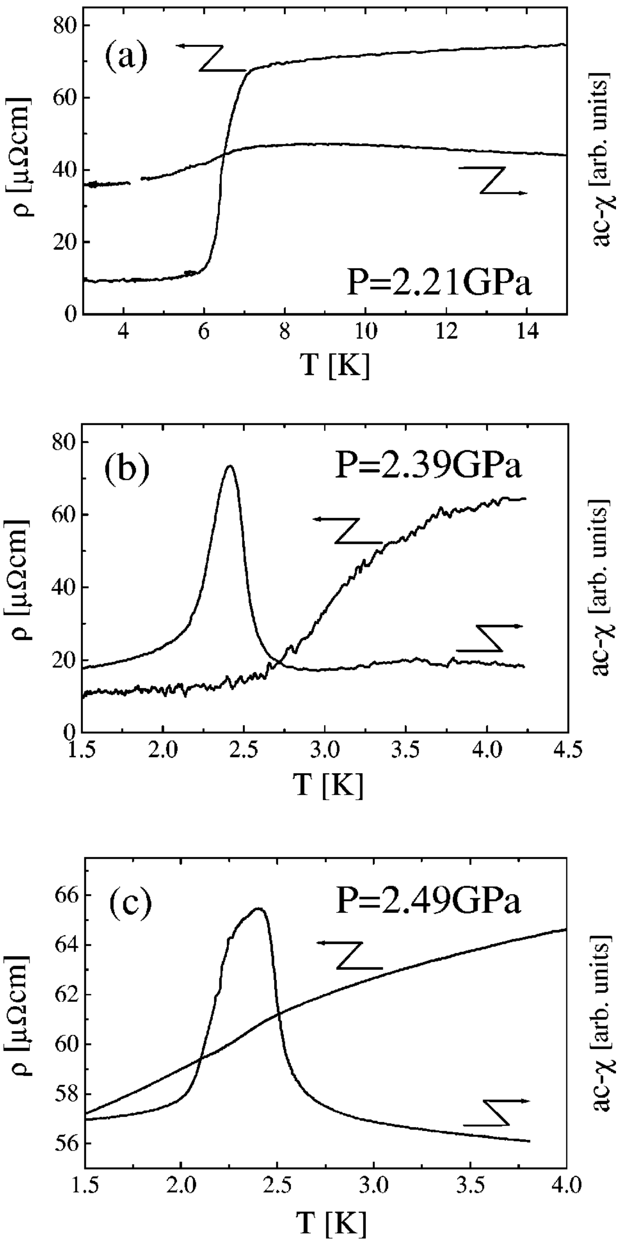

FIG. 3. Temperature dependence of the electrical resistivity and ac susceptibility of YbInCu_4 at (a) 2.21 GPa, (b) 2.39 GPa, and (c) 2.49 GPa.

mentioned above. We also observed that the magnitude of the χ_{ac} peak is strongly suppressed by applying low magnetic fields H, and almost disappears for $H>500$ Oe (see the inset of Fig. 4). These characteristic behaviors of χ_{ac} observed above 2.49 GPa are consistent with the onset of long-range ferromagnetic ordering.[17] Thus, we may conclude that the pressure-stabilized HT phase of YbInCu_4 most likely possesses a ferromagnetically ordered ground state. This is compatible with the occurrence of the weak ferromagnetism observed at 0.8 GPa for the Y-substituted compound $\text{Yb}_{0.8}\text{Y}_{0.2}\text{InCu}_4$ below 1.7 K.[15] However, with the χ_{ac} data alone, we cannot exclude a canted antiferromagnetic state as one of possible ground states. In order to study the magnetic structure, magnitude of moments, and mechanism of magnetic ordering, further experiments with neutron scattering and/or nuclear magnetic resonance measurements under pressure are required.

Figure 3(b) shows typical χ_{ac} and ρ data observed in the intermediate pressure region (2.28–2.44 GPa) between the valence transition and the magnetically ordered states. With decreasing temperature, we observed first a somewhat broadened step decrease around 3.5 K in ρ with a very small χ_{ac} decrease, which are attributed to the pressure-suppressed valence transition. Then, at lower temperature around 2.4 K, we observed the clear χ_{ac} peak, that can be ascribed to the long-range magnetic ordering. Here, one can clearly see that the χ_{ac} peak at T_M is much larger in magnitude than the decrease in χ_{ac} at T_V. The observation of both the magnetic ordering and the valence transition at distinct temperatures is thought to be due to pressure inhomogeneity or perhaps coexistence of the valence collapse and magnetic order. What we may conclude here is that the critical pressure between the valence fluctuation collapse and the magnetic ordering is located around 2.4 GPa.

The present experimental results are summarized in a temperature-pressure phase diagram for YbInCu_4 as depicted in Fig. 4. With increasing pressure, the valence fluctuation is suppressed and the magnetic HT phase is stabilized. For the pressure above $\sim$2.45 GPa, the valence transition almost disappears and the long-range magnetic ordering with ferromagnetic components is induced. The given phase diagram suggests a first-order-like transition between two phases at the critical pressure $\sim$2.45 GPa. The magnetic ordering temperature $T_M=2.4$ K is nearly independent of pressure in the range between 2.39 and 2.58 GPa.

Figure 2(b) shows ρ vs T^2 plot at 2.49 and 2.28 GPa which are just above and below the critical pressure. They are roughly proportional to T^2 below T_M (for 2.49 GPa) and $\sim$3 K (for 2.28 GPa). We tentatively fit the data with the formula expected in the Fermi liquid regime $\rho(T)=\Delta\rho_0 +AT^2$, where $\Delta\rho_0$ is the residual resistivity and A is the coefficient of the quadratic term. A is estimated as 0.03 (1.0) $\mu\Omega\,\text{cm/K}^2$ for 2.28 (2.49) GPa, which appears to change more steeply around the critical pressure than the previous report.[20] Although a contribution of spin waves to the A term which generally gives $T^n(n>2)$ may not be negligible just below T_M, such a large enhancement of the A value in the ferromagnetically ordered phase may suggest a formation of the strongly correlated heavy fermion particles. $\Delta\rho_0$ at 2.49 GPa is about five times larger in magnitude than the value for 2.28 GPa. The pressure-enhanced A and $\Delta\rho_0$ lead us to expect an occurrence of superconductivity near or in the ferromagnetic ordered phase in analogy with UGe_2 (Ref. 18) and URhGe.[19] For this reason, we measured ρ at 2.28 and 2.49 GPa down to $\sim$80 mK. As shown in the inset of Fig. 1, however, ρ did not show any sign of superconducting transition.

In conclusion, we have carried out electrical resistivity and ac susceptibility measurements for YbInCu_4 at pressures up to 2.58 GPa and temperatures down to $\sim$80 mK. The valence fluctuations are suppressed by applying pressure and completely disappear for the pressures above 2.49 GPa. In the ac-susceptibility data above 2.39 GPa, we first found the

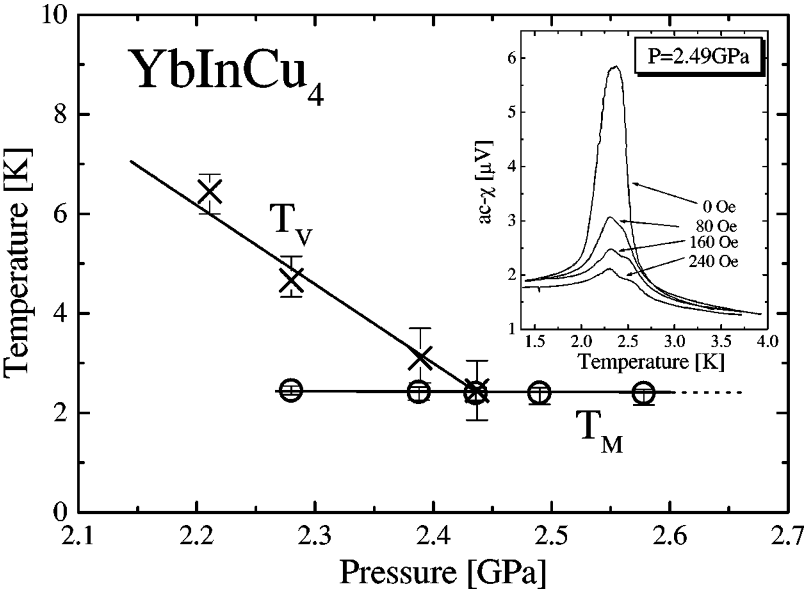

FIG. 4. The temperature-pressure phase diagram for YbInCu$_4$. The valence transition temperature T_V (cross) was defined at the midpoint of the transition. The vertical line for T_V shows the transition width from 10 to 90 % of the full transition. The magnetic transition temperature T_M (open circle) was defined at the maximum intensity of ac-susceptibility peak. The vertical line for T_M shows the full width of half maximum. The solid lines are guides to the eye. The inset shows the magnetic field dependence of the ac susceptibility at 2.49 GPa.

clear peak at around $T_M=2.4$ K with the small kink in the electrical resistivity, that can be ascribed to the onset of a long-range ferromagnetic ordering. Then we concluded that the ground state of the pressure-stabilized HT phase is most probably the ferromagnetically ordered state, though the possibility of a spin canted antiferromagnetically ordered state cannot be excluded at present. The critical pressure of ~2.45 GPa between two phases at low tempearture is easily obtainable with conventional techniques, and the present finding of magnetically ordered state will shed light on the peculiar magnetism in the Yb-based intermetallic compounds.

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