

## Summary of thesis:

# Ferromagnetic critical behavior and critical universality in itinerant-electron metamagnet UCoAl

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In the thesis, I report the static and dynamic properties of ferromagnetic (FM) critical phenomena observed in itinerant-electron metamagnet UCoAl.

### 1. Introduction

Itinerant-electron FM compounds have attracted much interest since novel phenomena, such as the coexistence of the FM state and the superconductivity in UGe<sub>2</sub>, were reported. Interestingly, these FM compounds are expected to follow an universal 3D (temperature - magnetic field - pressure) phase diagram characterized by the presence of a tricritical point (TCP), where a FM transition changes from second-order to first-order, and a critical endpoint (CEP), where a first-order field-induced FM transition [metamagnetic (MM) transition] changes to crossover. However, critical behaviors around these critical points have not been well studied due to a lack of suitable compounds. We focused on an itinerant-electron metamagnet UCoAl as a suitable example to clarify static and dynamic magnetic properties around these critical points since it shows the MM behavior at low magnetic fields along the *c*-axis  $\mu_0 H_{\parallel c} \sim 1$  T.

### 2. Experiment

We have performed nuclear magnetic resonance (NMR) and nuclear quadrupole resonance (NQR) measurements at <sup>27</sup>Al and <sup>59</sup>Co nuclear sites in UCoAl, and estimated *c*-axis magnetization ( $M_c$ ) (order parameter) from Knight shift ( $K$ ) measurements, and *c*-axis magnetic fluctuations ( $S_c$ ) from nuclear spin-lattice relaxation rate ( $1/T_1$ ) and nuclear spin-spin relaxation rate ( $1/T_2$ ) measurements. These measurements were performed under the precise control of tuning parameters: temperature ( $T$ ), magnetic field along the *c*-axis ( $H_{\parallel c}$ ), uniaxial pressure along the *b*-axis ( $P_{\parallel b}$ ) and the *c*-axis ( $P_{\parallel c}$ ), and substituted Fe concentration ( $x$ ).

### 3. Results and Discussions

#### 3-1. At ambient pressure

We observed the Ising-type anisotropy along the *c*-axis in both magnetization ( $M_c \gg M_a$ ) and magnetic fluctuations ( $S_c \gg S_a$ ). The longitudinal magnetic fluctuations well scale with the bulk magnetic susceptibility [ $S_c \propto \chi_c$ ], indicating that 3D-FM fluctuations are dominated in UCoAl. Furthermore,  $S_c$  exhibits a broad maximum at  $T_{\max} \sim 20$  K even in  $\mu_0 H_{\parallel c} = 0$ , which is a characteristic feature in itinerant-electron metamagnets.

At low temperatures, with increasing  $H_{\parallel c}$ , the coexistence of the paramagnetic (PM) and FM spectra, which is the characteristics of the first-order MM transition, was observed at  $\mu_0 H_{\parallel c} \sim 0.6$  T. At high temperatures, on the other hand, the continuous shift of spectra due to the crossover was observed. The contour plots of  $M_c$  and  $S_c$  on the  $T - H_{\parallel c}$  phase diagram at ambient pressure show the presence of the CEP at  $(T, \mu_0 H_{\parallel c}) \sim (12 \text{ K}, 1.0 \text{ T})$  and the divergence of  $S_c$  at the CEP. In addition, from the estimation of critical exponents ( $\delta, \beta, \gamma$ ) around the CEP, we found that the critical behaviors of the MM transition in UCoAl is categorized into the 3D-Ising universality class as observed in the gas-liquid and 3D-Mott transitions.

### 3-2. Uniaxial pressure and Fe-substitution effects

Uniaxial pressure  $P_{\parallel b}$  and  $P_{\parallel c}$  give rise to the opposite magnetic response in UCoAl.  $P_{\parallel b}$  increases the MM transition field  $H_{\parallel c}$  (i.e. suppresses the FM state), whereas,  $P_{\parallel c}$  induces the FM state even in  $\mu_0 H_{\parallel c} = 0$ . Furthermore, the peak intensity of  $S_c$  at  $T_{\max} \sim 20$  K in  $\mu_0 H_{\parallel c} = 0$  is suppressed by  $P_{\parallel b}$ , whereas, it is enhanced by  $P_{\parallel c}$  and maximized around  $(T, P_{\parallel c}) \sim (20 \text{ K}, 0.16 \text{ GPa})$ , indicating that FM fluctuations are enhanced toward the TCP.

In addition, Fe-substituted  $\text{U}(\text{Co}_{1-x}\text{Fe}_x)\text{Al}$  also shows the FM transition even in  $\mu_0 H_{\parallel c} = 0$  with a tiny Fe-concentration ( $x \geq 1\%$ ). The FM transition is first-order because the PM and FM spectra coexist without the continuous connection between them. The first-order FM transition line and  $T_{\max}$  line seem to merge around  $(T, x) \sim (20 \text{ K}, 2.5\%)$ , which suggests the presence of the TCP as theoretically predicted. Taking the above  $P_{\parallel c}$  measurement into consideration,  $S_c$  at  $T_{\max} \sim 20$  K develops with approaching the TCP.

## 4. Conclusion

From my Ph.D studies, I have confirmed the presence of the universal 3D phase diagram in the itinerant-electron metamagnet UCoAl and revealed the static and dynamic FM critical behaviors on the phase diagram. We found that UCoAl possesses Ising-type longitudinal FM fluctuations along the  $c$ -axis, which are maximized at a characteristic temperature  $T_{\max} \sim 20$  K. The enhanced FM fluctuations at  $T_{\max}$  are developed toward the critical endpoints of first-order MM/FM transitions (CEP/TCP) under the control of tuning parameters. We point out that the development of FM fluctuations around these critical points in the universal 3D phase diagram play an important role for exotic phenomena, such as the coexistence of the FM state and the superconductivity observed in UGe<sub>2</sub> and the field-induced superconductivity observed in URhGe.