Verification of Valence Transition in the Heavy Fermion $YbCo_2Zn_{20}$ by Volume Change

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Cubic $YbCo_2Zn_{20}$ has attracted interest for its "super" heavy electron state, as it possesses the giant electronic specific heat coefficient of 7900 mJ/K^2mol [1]. In $(Yb_{1-x}Sc_x)Co_2Zn_{20}$, where Sc is substituted for Yb, a jump in lattice constant was observed at x ~ 0.3 in powder X-ray diffraction experiments at room temperature [2]. Since the ionic radius of Yb^{3+} is smaller than that of Yb^{2+}, this reduction in lattice constant may indicate the stabilization of Yb^{3+} due to a valence crossover/transition. In this case, a magnetic order is expected to be observed for $x > 0.3$. However, no signs of a magnetic order have been detected down to at least 2 K. Therefore, this lattice anomaly might represent an unconventional quantum phase transition from non-magnetic valence fluctuated state of Yb^{2+}-Yb^{3+} for $x < 0.3$ to a non-magnetic Kondo lattice state where Yb^{3+} is almost stabilized (for $x > 0.3$.

In this study, neutron powder diffraction experiments were conducted on $(Yb_{1-x}Sc_x)Co_2Zn_{20}$ to detect lattice contraction due to the phase transition. As mentioned above, a decrease in lattice constant was observed at room temperature for $x \sim 0.3$. Since the ionic sizes of Yb^{3+} and Yb^{2+} are different, if a valence transition occurs, a significant change in the lattice constant is expected. While synchrotron X-ray diffraction is a powerful tool to observe lattice changes, a valence transition is expected to show more pronounced changes at lower temperatures. Therefore, we performed neutron diffraction experiments in this time.

Single crystal samples were synthesized by the Zn self-flux method with a mixture ratio of x = 0.1, 0.2, 0.3, 0.4, 0.5, and 0.6. Approximately 1 gram of each was powdered for the neutron diffraction experiments. The Yb and Sc compositions were determined by ICP emission analysis performed by Kojundokagaku Co., Ltd. on a portion of the powder samples prepared for the neutron scattering experiments. The determined compositions were x = 0.097, 0.193, 0.323, 0.445, 0.540, and 0.650. Each sample of these six compositions was sealed in vanadium cans, and neutron powder diffraction experiments were performed over the temperature range of 4.6 K to 280 K using the HERMES spectrometer and a GM cryocooler at the Japan Atomic Energy Agency.

The $CeCr_2Al_{20}$-type cubic crystal structure was confirmed for all samples. Rietveld analysis was performed on the obtained powder diffraction data using the FullProf analysis program to determine the lattice constants. As a result, the lattice constants decreased from 280 K toward lower temperatures for all compositions x. Additionally, in the composition x dependence at 4.6 K, the lattice constants decreased as x increased. Around x = 0.4, a deviation from the monotonic decrease was observed. This behavior roughly corresponds to the composition at which a jump in lattice constant was found in a previous study [2]. This lattice anomaly around x = 0.4 observed in our neutron diffraction experiments may reflect the valence crossover of Yb.

[1] M. S. Torikachvili *et al.*, Proc. Natl. Acad. Sci. U.S.A. **104**, 9960 (2007).
[2] N. Pouse, PhD thesis, Univ. of California (2018).

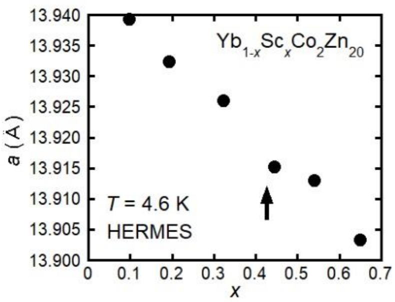

Fig. 1. Composition x dependence of lattice parameter in cubic $(Yb_{1-x}Sc_x)Co_2Zn_{20}$ at 4.6 K.