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 <b>MLF Experimental Report</b>	提出日 Date of Report 30 March 2015
課題番号 Project No. 2014A0187 実験課題名 Title of experiment μSR study of the magnetism and superconductivity in H-substituted Fe-based superconductor CeFeAsO <sub>1-x</sub> H <sub>x</sub> 実験責任者名 Name of principal investigator Masatoshi Hiraishi 所属 Affiliation Institute of Materials Structure Science, KEK	装置責任者 Name of responsible person Yasuhiro Miyake 装置名 Name of Instrument/(BL No.) MUSE D1 実施日 Date of Experiment 22-26 June 2014

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。(適宜、図表添付のこと)  
 Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.												
<table border="1" data-bbox="204 1037 871 1290"> <thead> <tr> <th>Chemical formulas</th> <th>Physical form.</th> <th>Mass / g</th> </tr> </thead> <tbody> <tr> <td>CeFeAsO<sub>0.62</sub>H<sub>0.54</sub></td> <td>Pelletized polycrystal</td> <td>1.0</td> </tr> <tr> <td>CeFeAsO<sub>0.36</sub>H<sub>0.62</sub></td> <td>Pelletized polycrystal</td> <td>1.0</td> </tr> <tr> <td>CeFeAsO<sub>0.34</sub>H<sub>0.66</sub></td> <td>Pelletized polycrystal</td> <td>1.0</td> </tr> </tbody> </table>	Chemical formulas	Physical form.	Mass / g	CeFeAsO <sub>0.62</sub> H <sub>0.54</sub>	Pelletized polycrystal	1.0	CeFeAsO <sub>0.36</sub> H <sub>0.62</sub>	Pelletized polycrystal	1.0	CeFeAsO <sub>0.34</sub> H <sub>0.66</sub>	Pelletized polycrystal	1.0
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2. 実験方法及び結果 (実験がうまくいかなかった場合、その理由を記述してください。) Experimental method and results. If you failed to conduct experiment as planned, please describe reasons.
<p>We performed muon spin relaxation (μSR) measurements at MUSE D1 area of MLF J-PARC on CeFeAsO<sub>1-x</sub>H<sub>x</sub> (Ce1111-H) with <math>x = 0.54, 0.62, \text{ and } 0.66</math> to investigate the magnetic ground state of the highly doped region. A standard zero field (ZF) μSR method using intense surface muon beam under the 300kW MLF beam power was conducted. Pellet shaped polycrystalline samples were mounted on the 15 x 15 or 20 x 20 mm<sup>2</sup> Ag sample holder, which is cooled by thermal conduction from the cryostat.</p> <p>It is revealed that ZF-μSR time spectra of Ce1111-H with <math>x = 0.54, 0.62, 0.66</math> below 100 K exhibit apparent reduction of the initial asymmetry <math>A_0</math> compared to the spectrum above 100 K, as shown in Fig. 1 (a, b, c), indicating that spatial segregation of non-magnetic and magnetic phases occurs with decreasing temperature. Due to the limitation of the instrumental time resolution, fast relaxation/rotation signal from the magnetic phase results in disappearance of the <math>A_0</math>. Moreover, spectra above magnetic transition temperature (<math>T_N</math>) exhibit <math>x</math>-independent exponential depolarization, which is currently attributed to Lorentzian field distribution due to a residual iron impurity, which is known to exist in the present samples. Considering these, we used the following</p>

## 2. 実験方法及び結果(つづき) Experimental method and results (continued)

form for the curve-fit analysis of the ZF- $\mu$ SR time spectra:  $A(t) = A_0 \exp(-\lambda t) + A_{\text{BG}}$ , where  $\lambda$  is the relaxation rate of the non-magnetic phase,  $A_0$  is a initial asymmetry for sample volume, and  $A_{\text{BG}}$  is a temperature independent background coming from muons stopped at Ag sample holder. Figure 2 shows the temperature dependence of the magnetically ordered volume fraction (MVF) in  $x = 0.54$  deduced from the  $A_0$ , where the magnetic phase emerges below  $\sim 100$  K, and gradually develops with decreasing temperature. MVF becomes mostly independent of temperature below  $\sim 50$  K to reach 0.84(2) at the lowest temperature. We determined the onset of  $T_N = 98(5)$  K by linear extrapolation, in the same manner, 100(8) K, 95(2) K, with  $x = 0.62, 0.66$ , respectively. Thus, it is revealed that  $T_N$  in  $x > 0.5$  is slightly lower than that in  $\text{LaFeAsO}_{1-x}\text{H}_x$  (La1111-H) with  $x > 0.5$ , as revealed by our previous experiment (proposal No. 2014AUD101).

To investigate the more detailed property of the magnetism, we performed additional  $\mu$ SR experiment using high-time resolution setup at PSI, Swiss with  $x = 0.44$  (non-superconducting sample). We observed the fast relaxation and oscillation of the signal below 80 K. It is revealed that rotating frequency at the lowest temperature [58(1) MHz] has much increased compared to that of  $x = 0.45$  in La1111-H [5.13(7) MHz], indicating that the magnetic structure of Fe atoms maybe different from one another. Magnetic volume fraction (MVF) at the lowest temperature with  $x > 0.44$  are almost unity, which is almost identical to that in La1111-H with  $x \geq 0.45$ . Additionally, we found that  $x$  dependence of the  $T_N$  seems to have a maximum around  $x = 0.62$ . Although this behavior is also analogous to La1111-H, maximum of the  $T_N \sim 100$  K is slightly lower than that of La1111-H ( $\sim 120$  K).

It is known that substitution of Ce for La brings in a chemical pressure to the system, resulting the decrease of the As-Fe-As bond angle  $\alpha$  and the increase of pnictogen height ( $h_{\text{As}}$ ). Since both are key parameters for the Fermi surface as pointed out by theoretical works, information on crystal structure in low-temperature phase is important for better understanding of the magnetism in this system. To achieve this, further investigation is currently in progress.

In summery,  $\mu$ SR experiment on Ce1111-H with  $x \geq 0.44$  reveals the existence of magnetic phase. There are difference of the magnetic properties between Ce1111-H and La1111-H, which may originate from the magnitude of the chemical pressure.

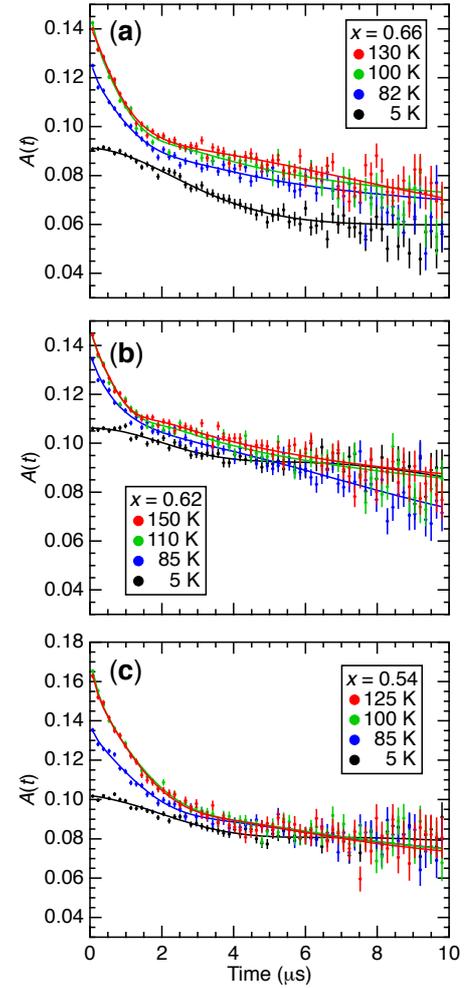


Figure 1: ZF- $\mu$ SR time spectra for  $x = 0.66$  (a),  $0.62$  (b), and  $0.54$  (c).

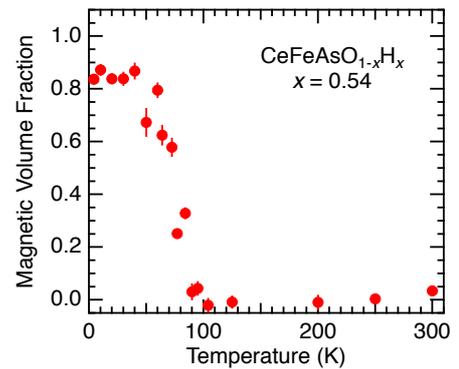


Figure 2: Fractional yield of magnetic phase in  $x = 0.54$ .