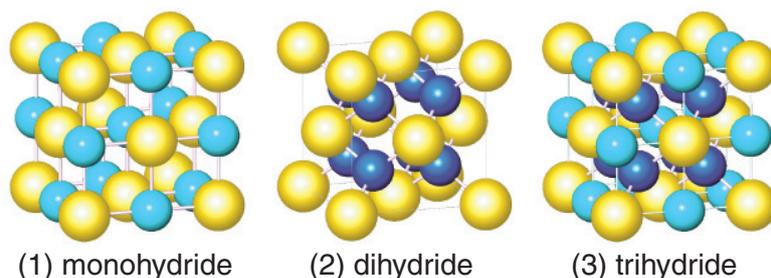
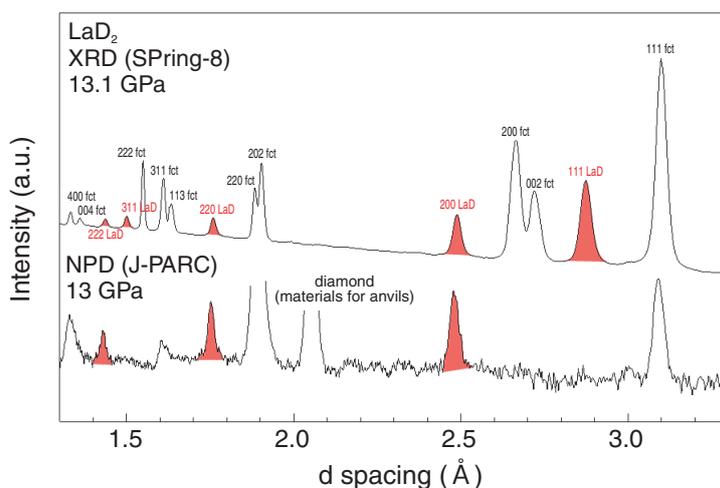


## 5-5 Discovery of Novel Rare-Earth Metal Hydrides under High Pressure — Formation of NaCl-Type Monohydrides by Pressure-Induced Phase Separation —



**Fig.5-12 Structures of three hydrides with different H concentrations and the fcc metal lattice**

Yellow, light-blue, and dark-blue spheres indicate metal atoms, H atoms at O sites, and H atoms at T sites, respectively. (1) Monohydride with only the O sites occupied; (2) dihydride with only the T sites occupied; and (3) trihydride with both O and T sites occupied.



**Fig.5-13 SR XRD (upper) and NPD (lower) profiles after phase separation of  $\text{LaD}_2$  at 13 GPa**

Intensities are on a logarithmic scale in arbitrary units. The reflection peaks of  $\text{LaD}$  are shown in red.

Rare-earth metals have a very high affinity for hydrogen (H) and form various H-concentration states, starting from a solid solution and ending with the trihydride  $\text{RH}_3$ , through the dihydride  $\text{RH}_2$ . Ideally,  $\text{RH}_2$  has a face-centered-cubic (fcc) metal lattice in which the tetrahedral (T) sites are filled with H atoms and the octahedral (O) sites remain empty (Fig.5-12(2)). Further absorbed H atoms occupy the O sites, and  $\text{RH}_3$  is eventually formed (Fig.5-12(3)). However, rare-earth metal monohydrides  $\text{RH}$  (Fig.5-12(1)), in which only the O sites are occupied by H atoms, has not yet been reported.

Previously, through synchrotron radiation (SR) X-ray diffraction (XRD) measurements at BL22XU, SPring-8, we found that  $\text{LaH}_2$  decomposed into two different fcc phases at pressures above 11 GPa. The observed decomposition suggests that H-poor and H-rich phases are spontaneously formed by pressurization. To understand this phase separation, information on the interstitial H atoms is necessary.

In this study, we attempted to determine the concentration and atomic positions of deuterium (D) after phase separation of  $\text{LaD}_2$  by neutron powder diffraction (NPD) at high pressure. First, we developed a high-pressure setup with a Paris-Edinburgh press and then installed it in the high-

intensity total diffractometer NOVA at BL21 in the MLF, J-PARC. With this setup, we succeeded in measuring NPD patterns up to 17 GPa, the highest pressure for domestic NPD experiments, at ambient temperature.

Fig.5-13 shows the XRD and NPD profiles obtained at ~13 GPa. The Bragg peaks of the D-poor phase with an fcc metal lattice are shown in red. One may notice that several Bragg peaks indexed with odd numbers are missing in the NPD pattern for the D-poor phase, while these peaks are present in the XRD pattern. The lack of odd-number-indexed Bragg peaks suggests the formation of an NaCl-type structure due to the comparable neutron scattering length of the La and D atoms. The NaCl-type hydride can be regarded as an fcc monohydride, in which only the O sites are occupied by H(D) atoms. The present result indicates that only rare-earth metals can form a series of stoichiometric hydrides, such as  $\text{RH}$ ,  $\text{RH}_2$ , and  $\text{RH}_3$ , with the fcc metal lattice.

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### Reference

Machida, A. et al., Formation of NaCl-Type Monodeuteride  $\text{LaD}$  by the Disproportionation Reaction of  $\text{LaD}_2$ , Physical Review Letters, vol.108, issue 20, 2012, p.205501-1-205501-5.