

Anomalous Structural Behavior in the Magnetic and Structural Transition of FeRh Thin Films from a Local Viewpoint

The antiferromagnetic-to-ferromagnetic (AFM-to-FM) transition of FeRh thin films was investigated by Fe and Rh *K*-edge X-ray absorption fine structure spectroscopy (XAFS) in order to clarify the correlation between its global magnetism and local electronic/geometrical structures. A strong Fe-Rh hybridization, which decreases from AFM to FM phases, was revealed. Moreover, only the Fe-Fe Debye-Waller factor in the AFM phase was observed to be enhanced in comparison with that in the FM phase. This behavior suggests the importance of the interplay between the local spin and Fe-Fe distance fluctuations near the phase transition.

Recently, CsCl-type ordered FeRh alloy near the equiatomic stoichiometry is attracting renewed interest in view of its potential application to various magnetic devices. Upon increasing its temperature, it shows an antiferromagnetic-to-ferromagnetic (AFM-to-FM) phase transition accompanied with a nearly 1% volume expansion at ~350 K [1]. Owing to this magnetic and structural phase transition occurring at mild temperature, FeRh is a promising material for thermally-assisted magneto-recording, antiferromagnetic memory, and electric-field-induced magnetism switching [2-4]. Therefore, it is important to elucidate the origin of the phase transition in FeRh not only from the viewpoint of basic research, but also for device development. Despite many studies on its phase transition, the physical origin and the essential interactions involved are still hot topics. Especially, the relationship between spin, structure, and electronic state, which all change through the phase transition, is a key issue. In this study, FeRh thin film was investigated by using X-ray absorption fine structure (XAFS) spectroscopy. XAFS spectroscopy is a powerful method for

detecting both atomic-dependent and bond-dependent local structures. According to the change in the X-ray absorption near-edge structure (XANES) and Debye-Waller factors of Fe-Rh and Fe-Fe, we consider that the local Fe-Fe distance and the spin fluctuations act as precursors. These fluctuations play an important role in driving the phase transition.

100-nm-thick Fe₄₉Rh₅₁ thin film was epitaxially grown on a single crystal MgO(001) substrate by magnetron co-sputtering of Fe and Rh targets. The occurrence of AFM-to-FM phase transition at 385 K, consistent with the previously known transition temperature of FeRh/MgO(001) thin film, was verified by using a superconducting quantum interference device (SQUID, Quantum Design MPMS-7) DC magnetometer at 300 Oe and by using the longitudinal magnetic optical Kerr effect. Further details of the sample preparation and evaluations are described in reference [5]. The XAFS spectra for Fe and Rh *K* edges were obtained at beamlines BL-9A and AR-NW10A, respectively, in fluorescence yield mode by using a multi-element Ge solid state detector.

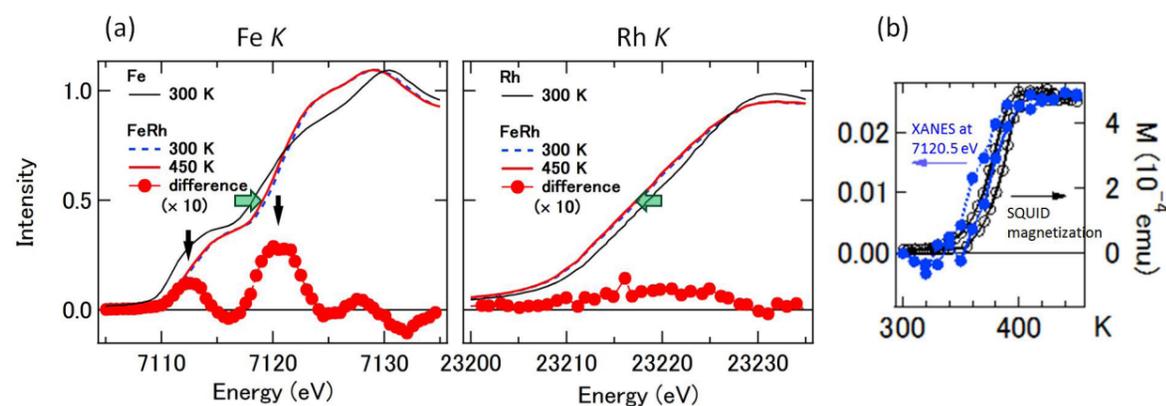


Figure 1: (a) 300 K (AFM) and 450 K (FM) XANES of Fe and Rh *K* edges of pure metals and FeRh. (b) Comparison of the XANES spectral intensity at 7120.5 eV (filled blue circles) with the magnetic moment *M* (open black circles).

Figure 1a shows the XANES spectra at Fe and Rh *K* edges of FeRh at 300 K (AFM) and 450 K (FM), respectively, together with those of pure Fe and Rh metal foils. Compared with pure foils, it can be seen that the Fe *K* edge of FeRh is shifted to higher energy, whereas the Rh *K* edge shifts oppositely. This suggests electron transfer from Fe to Rh due to their strong hybridization and their difference in electronegativity. Moving on to the difference spectra of FeRh between 450 K and 300 K in **Fig. 1a**, we could observe a slight yet distinct increase in the spectral weight at around 7112.5 and 7120.5 eV in the Fe *K*-edge XANES spectrum. The temperature evolution of the intensity of these peaks depicts hysteresis loops similar to the magnetization (**Fig. 1b**). The suppression of the Fe-Rh hybridization would result in the decrease of the spectral intensity at these energies across the phase transition.

Figure 2 shows the Debye-Waller factors C_2 near the transition temperature extracted from the Fe *K*-edge extended XAFS oscillations. The Debye-Waller factor of Fe-Rh exhibits a normal behavior even during the phase transition: increasing monotonically with temperature rise due to the enhancement of the usual thermal vibrations. In contrast, the Debye-Waller factor of Fe-Fe shows a decrease during heating from the AFM to FM phases in the phase separation region. Since the phase separation would give a maximum static fluctuation near the middle point of the transition, the present finding indicates that the maximum Fe-Fe distance fluctuation is due to the inherent nature of the AFM phase near the phase transition.

According to a recent theoretical study, the AFM exchange interaction between two nearest-neighbor Fe atoms depends more strongly on volume change than the FM exchange interaction between nearest Fe and Rh atoms [6]. The balance of these exchange interactions is expected to determine the global magnetic state. At a small volume the AFM state is exclusively favored, and the local spin fluctuation would be coupled with the Fe-Fe distance fluctuation. Thus, the anomalous behavior of the Fe-Fe Debye-Waller factor could be ascribed to the dynamic fluctuations of local distances and spins in the AFM phase near the phase transition. On the contrary, the FM state is expected to allow for the spin fluctuation more easily because of the weakening of the Fe-Fe AFM exchange interaction due to a longer Fe-Fe distance [6], resulting in a rather small Fe-Fe Debye-Waller factor. Therefore, the anomalous behavior found in the Fe-Fe Debye-Waller factor indicates that the local Fe-Fe distance and the spin fluctuations play an important role in driving the phase transition.

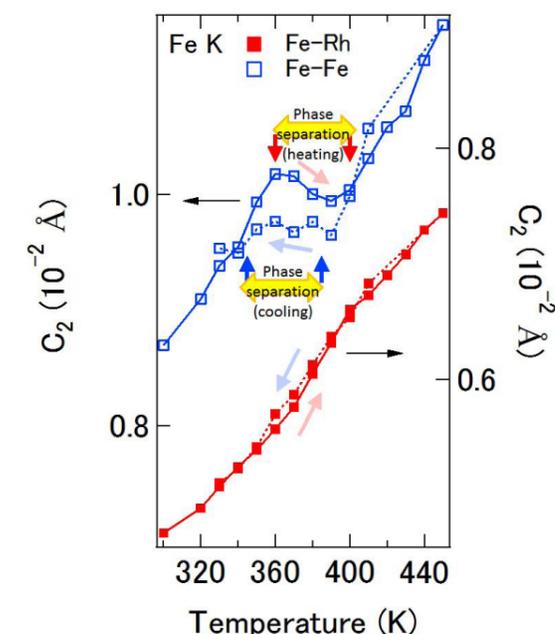


Figure 2: Enlarged view from 300 to 450 K for the Fe-Rh and Fe-Fe Debye-Waller factors extracted from Fe *K*-edge extended XAFS.

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BEAMLINES

BL-9A and AR-NW10A

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