

## Article

# Electric Field Control of Magnetic Properties by Means of Li<sup>+</sup> Migration in FeRh Thin Film

Gengfei Li<sup>1,2</sup>, Yali Xie<sup>1,2,\*</sup> , Baomin Wang<sup>1,2</sup> , Huali Yang<sup>1,2</sup> and Run-Wei Li<sup>1,2,3</sup>

- <sup>1</sup> CAS Key Laboratory of Magnetic Materials and Devices, Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences, Ningbo 315201, China; ligengfei@nimte.ac.cn (G.L.); wangbaomin@nimte.ac.cn (B.W.); yanghl@nimte.ac.cn (H.Y.); runweili@nimte.ac.cn (R.-W.L.)
- <sup>2</sup> Zhejiang Province Key Laboratory of Magnetic Materials and Application Technology, Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences, Ningbo 315201, China
- <sup>3</sup> Center of Materials Science and Optoelectronics Engineering, University of Chinese Academy of Sciences, Beijing 100049, China
- \* Correspondence: xieyl@nimte.ac.cn

**Abstract:** Recently, the electric control of magnetism by means of ion migration has been proven to be effective with nonvolatility and low energy consumption. In this work, we investigated the control of the magnetic properties of FeRh films by means of Li<sup>+</sup> migration in FeRh/MgO heterostructures. We found that the migration of Li<sup>+</sup> could reduce the phase transition temperature by 2 K with an applied voltage of 1 V. Meanwhile, the voltage-dependent saturated magnetization exhibited a repetitive switching behavior from high to low magnetization values while the voltage was switched from 4 to −4 V, indicating that the migration of Li<sup>+</sup> in the FeRh film can be reversible. This provides a means to control the magnetic properties of FeRh films.



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**Keywords:** electric control of magnetic phase transition; FeRh; Li<sup>+</sup> migration

## 1. Introduction

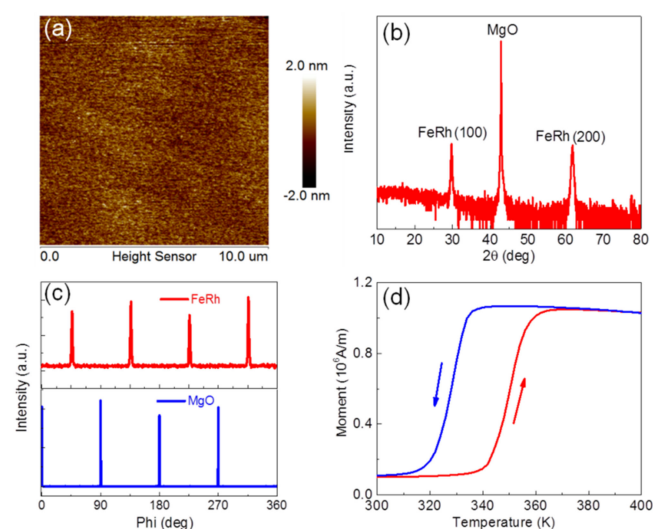
Fallot et al. discovered that the CsCl-type FeRh alloy undergoes a first-order phase transition from an antiferromagnetic (AF) to ferromagnetic (FM) phase at around room temperature accompanied by an approximately 1% volume expansion of the crystal lattice [1]. Due to this magnetic transition, FeRh has attracted extensive attention for its potential applications in heat-assisted magnetic recording, AF memory, and magnetic refrigeration [2–7]. In order to realize its applications under different conditions, many studies have been conducted to tune the magnetic properties of the FeRh film by means of the strain, magnetic field, electric field, etc. [8]. However, the manipulation of FeRh properties by means such as the strain and magnetic field is difficult to implement in spintronics devices, limiting its practical applications [9,10]. In recent years, the electric control of magnetism has been proven to be effective with low energy consumption and repeatability [11–16]. For example, Cherifi et al. found that a moderate electric field can produce a large magnetization variation in FeRh/BaTiO<sub>3</sub> [14]. Lee et al. reported a large resistivity modulation by applying an electric field to FeRh/PMN-PT heterostructures [16]. However, these methods usually require a high-quality interface or very thin film. Recently, the electric control of magnetism by means of ion migration has been proven to be effective with nonvolatility and low energy consumption [17–19]. For instance, Dhanapal et al. demonstrated reversibly controlled magnetic domains of Co via electric-field driven oxygen migration at the nanoscale [18]. In this work, we investigated the electric control of magnetic properties of FeRh films by means of Li<sup>+</sup> migration in FeRh/MgO heterostructures. We found that this method can not only manipulate the phase transition temperature, but also change the saturated magnetization of FeRh film.

## 2. Experiment

First, 30 nm thick FeRh films were deposited by sputtering from an FeRh alloy target using a DC power of 100 W and an argon pressure of 5 mTorr. The (100)-oriented MgO substrates were preheated to 530 °C for 1 h in a vacuum chamber and then held at this temperature during deposition. After growth, the samples were annealed at 700 °C for about 1 h at a base pressure below  $1.0 \times 10^{-5}$  Pa in order to obtain the chemically ordered CsCl-type FeRh. The film thicknesses were calibrated by X-ray reflectivity (XRR) and the rate of growth was 5 nm/min. The surface topographies of FeRh films were measured using a Bruker Icon atomic force microscope. The crystal structure was characterized using a Bruker Discover X-ray diffractometer. The temperature-dependent magnetizations of the FeRh films were determined using a superconducting quantum interference device (SQUID) magnetometer. The electric field was applied using a commercial three-electrode experimental setup [20]. In this work, FeRh/MgO, a standard calomel electrode, and a platinum sheet were used as the working electrode, reference electrode, and counter electrode, respectively. In addition, 0.1 M of LiClO<sub>4</sub> dissolved in propylene carbonate was used as the electrolytic solution. When voltage was applied, the Li<sup>+</sup> could migrate into or out of the FeRh films. When we performed magnetic measurements, the FeRh/MgO sample was taken out of the experimental setup and the electrode setup was also removed.

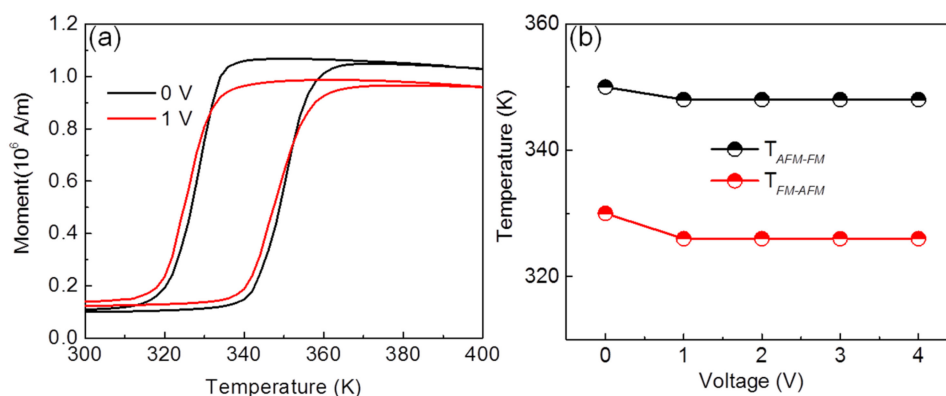
## 3. Results and Discussion

An AFM image of the FeRh films is displayed in Figure 1a, which indicates that the roughness of the film was very small ( $\sim 0.3$  nm). Figure 1b shows the XRD  $\theta$ - $2\theta$  pattern of the FeRh film. As seen in Figure 1b, only the reflections at 29.90° and 62.11° could be observed, which demonstrates that the obtained FeRh film had the required CsCl-type structure. The X-ray  $\varphi$ -scan of FeRh/MgO in Figure 1c indicates that the FeRh films were epitaxially grown with a 45° in-plane lattice rotation with respect to the MgO(001) substrates. The temperature-dependent magnetization of the FeRh film measured with an in-plane magnetic field of 2 kOe is plotted in Figure 1d. When warming up, the magnetization steeply increased from 100 to  $1.1 \times 10^6$  A/m at around 350 K, exhibiting a typical characteristic of the transition from the AF to the FM phase in FeRh. By contrast, the magnetization of the FeRh film gradually decreased to the original value of the AF state in the cooling process. A temperature hysteresis in the magnetization of about 25 K could be observed, indicating the first-order phase transition of FeRh.



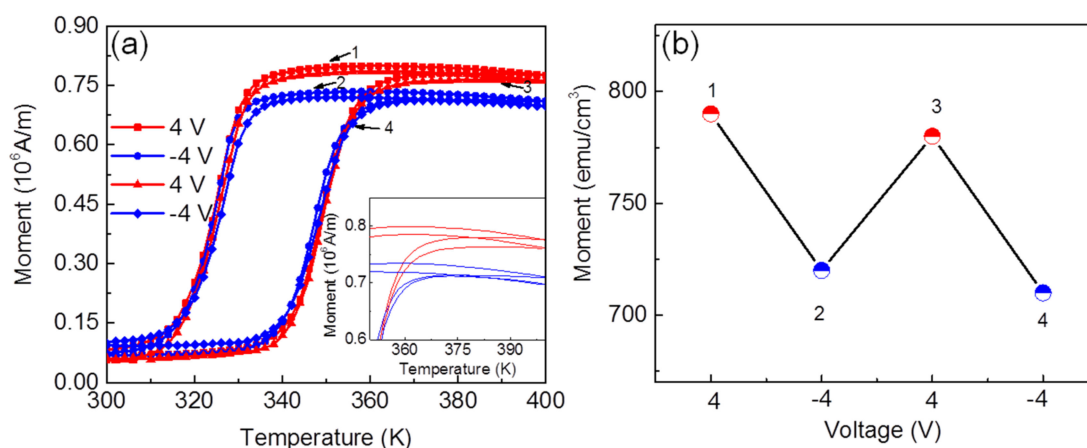
**Figure 1.** (a) The AFM image of the FeRh films. (b)  $\theta$ - $2\theta$  scan and (c)  $\varphi$ -scans of FeRh/MgO. (d) The temperature-dependent magnetization of the FeRh film measured with an in-plane magnetic field of 2 kOe.

Figure 2a shows the temperature-dependent magnetization of the FeRh film at 2 kOe without and with applied voltages of 1 V through the electrochemical method. It can be seen that with an applied voltage of 1 V, the behavior of the temperature-dependent magnetization was almost unchanged compared with that for 0 V. Meanwhile, the phase transition shifted to the lower temperature after the applied voltage of 1 V. The voltage-dependent critical phase transition temperatures of FeRh, which were defined as  $T_{AFM-FM}$  on heating and  $T_{FM-AFM}$  on cooling processes, are plotted in Figure 2b.  $T_{AFM-FM}$  exhibited a decrease of about 2 K with an applied voltage of 1 V, indicating that the  $Li^+$  migrated into the FeRh film. However, no further change was observed after increasing the applied voltages.  $T_{FM-AFM}$  behaved similarly to that of  $T_{AFM-FM}$ . This kind of behavior may be ascribed to the small size of  $Li^+$ . When voltage was applied, the  $Li^+$  could migrate into the FeRh film and the lattice volume could expand, which have been confirmed in our previous work [21]. The magnetization and phase transition temperature of FeRh are strongly dependent on the lattice parameter [22–24]. Consequently, the  $T_{AFM-FM}$  and  $T_{FM-AFM}$  change due to the migration of  $Li^+$ . However, the lattice volume cannot change by very much, due to the small size of  $Li^+$ , leading to a modulation of about 2 K in  $T_{AFM-FM}$  and  $T_{FM-AFM}$  when a voltage of 1 V was applied, as well as an easy saturation.



**Figure 2.** (a) Temperature-dependent magnetization without and with an applied voltage of 1 V for the FeRh thin film at 2 kOe. (b) Voltage-dependent  $T_{AFM-FM}$  and  $T_{FM-AFM}$  of FeRh on heating and cooling processes.

Figure 3a shows the reversibility of the temperature-dependent magnetization of the FeRh film measured with applied voltages of 4 and  $-4$  V. The applied magnetic field was 2 kOe. It can be seen that when 4 V voltages were applied, the saturated magnetization of the first temperature-dependent magnetization (1) at 400 K could reach about  $0.8 \times 10^6$  A/m. Meanwhile, when voltages of  $-4$  V were applied, the saturated magnetization of the first temperature-dependent magnetization (2) at 400 K decreased to about  $0.7 \times 10^6$  A/m, indicating that applying an electric field of  $-4$  V can lead to the decrease in the saturated magnetization compared with that for a applied voltages of 4 V. When applied voltages of 4 and  $-4$  V were applied again, the change in the saturated magnetization (3 and 4) was repeated. This kind of behavior can be seen more clearly in the inset of Figure 3a. It can also be ascribed to the migration of  $Li^+$ . When 4 V voltages were applied, the  $Li^+$  can migrate into the FeRh film. However, when voltages of  $-4$  V were applied, the  $Li^+$  migrated out of the FeRh film, leading to a decrease in the saturated magnetization. The saturated magnetizations under different voltages are summarized in Figure 3b. It can be seen that the voltage-dependent saturated magnetization exhibited a repetitive switching behavior from high to low magnetization values when switching the voltage from 4 to  $-4$  V repetitively, indicating that the migration of  $Li^+$  in FeRh film can be reversible.



**Figure 3.** (a) Temperature-dependent magnetization with applied voltages of 4 and  $-4$  V for FeRh thin films. The inset shows the enlarged figure of the temperature-dependent magnetization. (b) Voltage dependence of saturated magnetization measured with a magnetic field of 2 kOe in FeRh thin films.

#### 4. Conclusions

In summary, the magnetic properties of FeRh films can be controlled by means of electrochemistry in FeRh/MgO heterostructures. Both the phase transition temperature and saturated magnetization of FeRh films can be modulated by varying the voltage. This kind of modulated behavior can be ascribed to the migration of  $\text{Li}^+$  ions by applying voltage. This work provides a new method to control the magnetic properties of FeRh films.

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