

# Preparation and magnetic properties of wrinkled FeRh flexible films

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## ABSTRACT

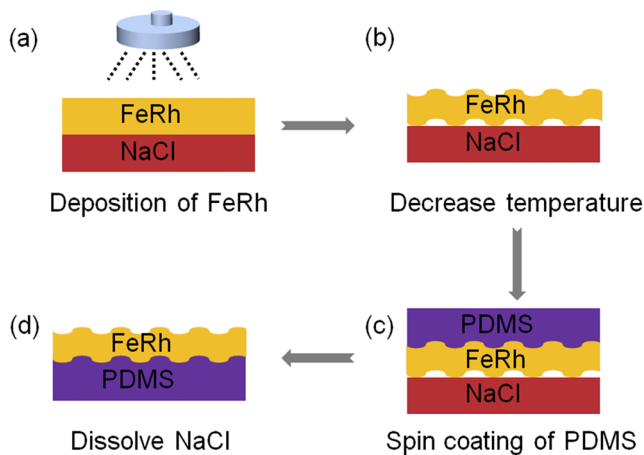
Magnetic thin films are indispensable in flexible devices, which necessitate methods to fabricate flexible magnetic thin films. In this work, we present a method to fabricate wrinkled FeRh flexible films via a coating and transfer process. The obtained FeRh/PDMS films have random patterns of wrinkles with a mean periodicity of 10  $\mu\text{m}$ . From the curvature dependent magnetic measurements, it can be determined that the magnetic properties of the obtained wrinkled FeRh flexible films are insensitive to flexing up to a radius of curvature of 1.5 mm, making it promising for applications in flexible devices.

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## I. INTRODUCTION

Recently, flexible device which is fabricated on plastic/polymer substrates have drawn people's attention because of the promise in applications such as smart cards, wearable electronics, sensors, etc.<sup>1–4</sup> Since magnetic layers are a key component of these flexible devices, flexible magnetic films have also attracted wide attention.<sup>5</sup> FeRh alloy which has CsCl-type structure have been used widely as one of the magnetic layer in magnetic devices, such as in magnetic recording, because of the first-order phase transition which changed from antiferromagnetic (AF) at room temperature to ferromagnetic (FM) phase upon heating above 300 K.<sup>6–8</sup> Due to this magnetic transition, FeRh has attracted extensive attention for its potential application in spintronic devices.<sup>9–14</sup> However, the growth temperature of FeRh thin films is too high for most plastic/polymer substrates, which is a challenge to produce flexible FeRh thin films.<sup>15</sup> On the other hand, the magnetic properties of FeRh can

be changed by applying strain to the flexible substrate due to inverse magnetostrictive effects and magnetoelastic coupling.<sup>16–20</sup> As a result, the original magnetic state will be changed. More importantly, the intrinsic elastic properties of FeRh thin film and plastic/polymer substrates have great disparities in this metal/plastic or metal/polymer architecture, which make it easy to crack due to the fragile nature of the FeRh thin film. In recent years, due to the repeatedly stretched and fully recovered nature, artificial wrinkling structures have been widely used in fabricating different stretchable electronics.<sup>21–31</sup> This kind of structure can be used to prepare the flexible magnetic films in which the magnetic properties are insensitive to the applied strain during bending or stretching. In this work, we present a method to fabricate wrinkled FeRh films via a coating and transfer process. Our results show that the fabricated FeRh flexible thin film exhibits nearly unchanged magnetic properties with curvature radii decreasing from  $\infty$  to 1.5 mm.



**FIG. 1.** [(a)–(d)] Step-by-step process for fabricating flexible FeRh/PDMS thin films with random wrinkles.

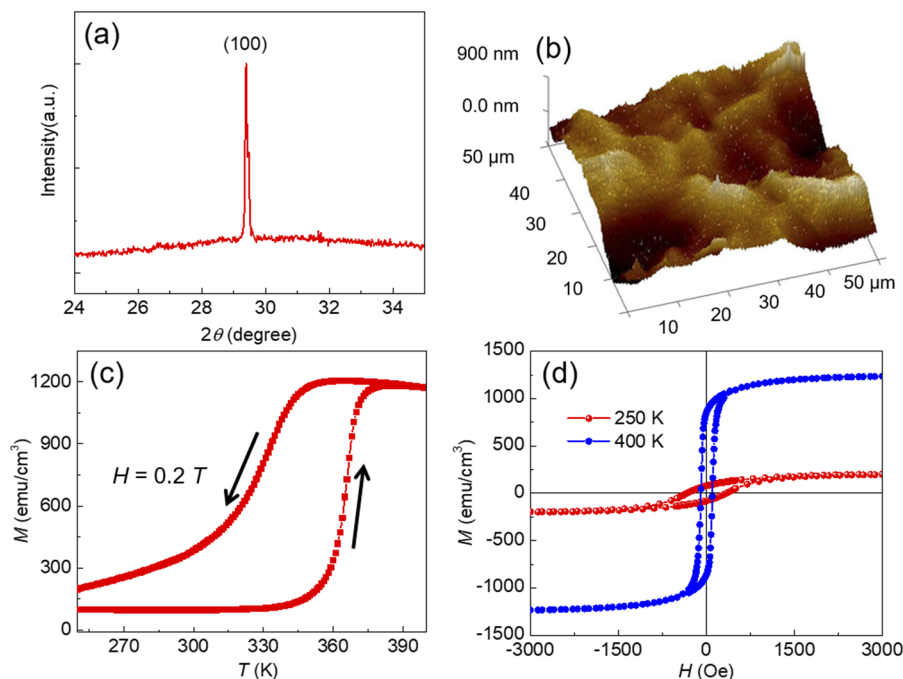
## II. EXPERIMENT

FeRh films were deposited by magnetron sputtering with a base pressure below  $9 \times 10^{-5}$  Pa. The used substrate was NaCl single crystal, as shown in Fig. 1(a). The film thickness is about 80 nm. Firstly, the substrates were annealed at 500 °C for 1 h in the vacuum chamber, then the temperature was held at 650 °C during deposition. After growth, FeRh films were annealed at 670 °C for 1 h and then cooled to room temperature. There are great disparities in the thermal expansion coefficient of FeRh and NaCl crystal substrates, e.g.,

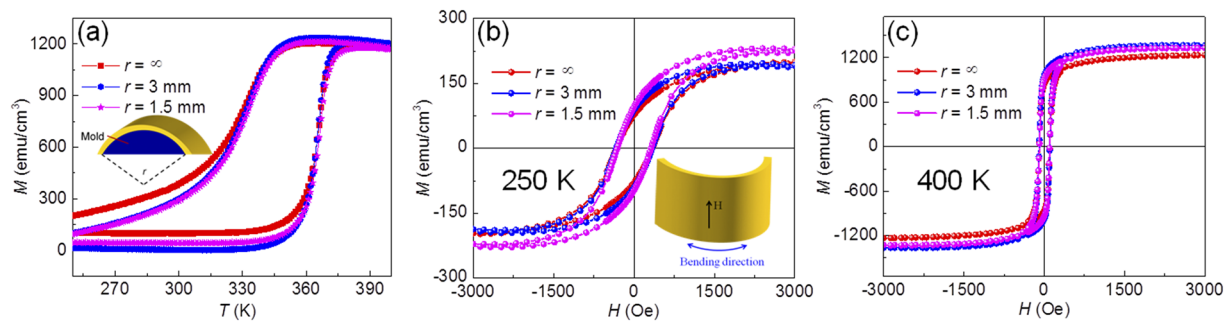
the thermal expansion coefficient is about  $12 \times 10^{-6} \text{ K}^{-1}$  for metals such as FeRh, but  $40 \times 10^{-6} \text{ K}^{-1}$  for NaCl. Consequently, the grown films display random wrinkles, as shown in Fig. 1(b). Polydimethylsiloxane (PDMS) with a thickness of about 50  $\mu\text{m}$  was coated on the top side of the wrinkled film surface using a spin coating technique, as schematically shown in Fig. 1(c). After the spin coating process, the NaCl crystal is removed from the FeRh films, by means of dissolution, leading to a PDMS elastomeric template covered with the FeRh film [Fig. 1(d)]. The film thicknesses were controlled by the deposition time, which have been calibrated by X-ray reflectivity. The crystal structure was characterized using a Bruker D8 Discover X-ray diffractometer with  $\text{CuK}\alpha$  radiation source. The  $2\theta$  range was 20–70° with a step size of 0.04°. The surface topography of the films was measured in a Bruker Icon atomic force microscope (AFM, Veeco Dimension 3100 V) using tapping mode. A Quantum Design superconducting quantum interference device-vibrating sample magnetometer (SQUID-VSM) was employed to measure the temperature dependent magnetization and hysteresis loops for the FeRh/PDMS in the temperature range from 250 to 400 K. The volume of the FeRh thin film was approximated to the value of the sample area multiple by its thickness. During testing, samples were fixed on molds with different curvature radii.

## III. RESULTS AND DISCUSSION

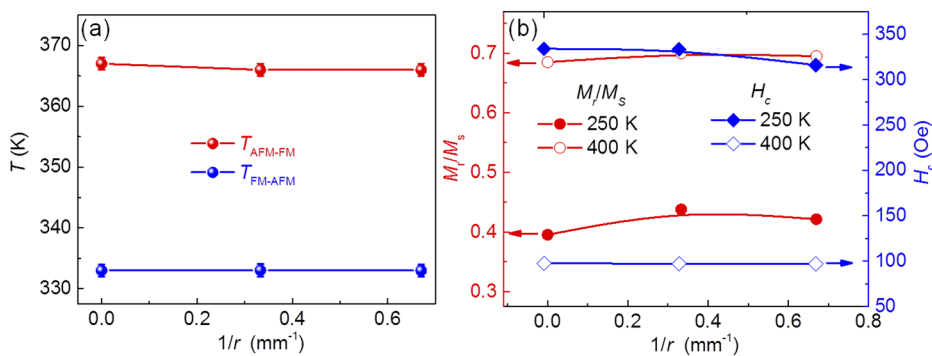
Figure 2(a) shows the XRD  $\theta$ – $2\theta$  pattern of the obtained FeRh/PDMS sample. Though the lattice mismatch between FeRh and NaCl is very large (–5.3%), only the (100) peak of the CsCl-type FeRh can be seen, which demonstrates the oriented growth of the film. The three-dimensional AFM topography of the FeRh/PDMS displayed in Fig. 2(b) was measured to make sure that the formed



**FIG. 2.** (a) XRD pattern of the FeRh/PDMS flexible thin film. (b) Three-dimensional plot of the AFM image. (c) Temperature dependent magnetization for FeRh/PDMS in the heating and cooling process at 0.2 T. (d) Typical hysteresis loops measured at 250 and 400K.



**FIG. 3.** (a) Temperature dependent magnetization for FeRh/PDMS under various radii of curvature in an applied magnetic field of 0.2 T. Typical hysteresis loops under various 0 radii of curvature are shown, measured at (b) 250 K and (c) 400 K for FeRh/PDMS.



**FIG. 4.** (a) The phase transition temperatures,  $T_{AF-FM}$  and  $T_{FM-AF}$  of FeRh/PDMS, measured at three different curvature amounts. (b) The coercive field,  $H_c$ , and normalized remanent magnetization,  $M_r/M_s$ , measured at  $T = 250$  and 400 K.

wrinkles were random in the FeRh thin film. A random pattern of wrinkles with a mean periodicity of 10  $\mu\text{m}$  is induced in the FeRh film by the removal of the rigid substrate. The roughness of the flexible FeRh thin film is as large as  $\sim 250$  nm. The temperature dependent magnetization and hysteresis loops of the obtained FeRh/PDMS were measured to characterize the magnetic properties of the wrinkled FeRh thin film, as shown in Figs. 2(c)–2(d). It can be seen that the magnetization is rather small at room temperature, and then gradually increases during the heating process, indicating the typical AF-FM phase transition of FeRh. Oppositely, the magnetization of the FeRh films gradually decreases during the cooling process. The temperature hysteresis of the magnetization is about 30 K due to the first order phase transition of the FeRh thin film. This kind of temperature dependent magnetization behavior has been reported in FeRh thin films grown on other types of rigid substrates.<sup>11</sup> The hysteresis loops of FeRh/PDMS measured at 250 and 400 K in Fig. 2(d) are also similar with that of FeRh films grown on other rigid substrates,<sup>10</sup> indicating that the obtained wrinkled FeRh thin films have similar magnetic properties with that of FeRh thin films grown on rigid substrates.

Figure 3(a) shows the temperature dependent magnetization of FeRh/PDMS under various levels of curvature in an applied magnetic field of 0.2 T. While decreasing the radius of curvature from  $\infty$  to 1.5 mm, the temperature dependent magnetic behaviors of FeRh/PDMS keep almost unchanged, indicating that the magnetic properties and the phase transition are stable even when the

sample is under various levels of curvature. Figure 3(b)–3(c) show the hysteresis loops of FeRh/PDMS with different amounts of bending, which were measured at 250 and 400 K, respectively. Similarly, the magnetic hysteresis loops are almost unchanged as the curvature decreased from  $\infty$  to 1.5 mm. Figure 4(a) summarizes the critical phase transition temperatures  $T_{AF-FM}$  and  $T_{FM-AF}$  of the FeRh/PDMS thin film. The transition temperatures ( $T_{AF-FM}$  and  $T_{FM-AF}$ ) are determined by the temperature with a maximum value in the first derivative of the temperature dependent magnetization curves. It shows that the  $T_{AF-FM}$  and  $T_{FM-AF}$  of the film is stable with radii of curvature decreasing from  $\infty$  to 1.5 mm, demonstrating that the critical phase transition temperatures are not affected by the bending of the film. Figure 4(b) displays the coercivity and  $M_r/M_s$  of FeRh/PDMS as a function of the radii of curvature at different temperatures. Similarly, the coercivity and  $M_r/M_s$  are almost constant when the curvature decreases from  $\infty$  to 1.5 mm. That is, the magnetic properties of wrinkled FeRh flexible films are insensitive to the bending moment applied to them. In fact, the FeRh flexible films did not suffer strain during bending due to the wrinkled structure. Consequently, there is no change in magnetic properties with increased bending curvatures.

#### IV. SUMMARY

We present a method to fabricate wrinkled FeRh flexible thin films via a coating and transfer process. A random pattern of

wrinkled FeRh thin films were obtained on flexible PDMS substrates following this method. The temperature dependent magnetization and hysteresis loops with different radii of curvature were measured to deduce any changes in magnetic transition temperature, relative remnant magnetization and coercivity. It can be concluded that the magnetic properties are insensitive to film curvature in wrinkled FeRh thin films. This kind of flexible magnetic thin film is suitable for applications on flexible devices.

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