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Magnetic properties and first-order magnetic phase transition in single crystal FeRh thin film

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Abstract To understand the mechanism of first-order magnetic phase transition in ordered FeRh thin films, the magnetic properties and first-order antiferromagnetic (AFM)–ferromagnetic (FM) phase transition behavior of single-crystal FeRh thin film are investigated in detail. The first-order magnetic phase transition is seen at a temperature of around 120 °C during heating and 145 °C cooling processes in perpendicular direction. The M–H loops measured isothermally amidst the AFM–FM transition regime show an opening at high magnetic field, which indicate a reversible AFM–FM transition induced by magnetic field. The clusters of the FM phase nucleate in the AFM matrix heterogeneously and vice versa during the first-order phase transition and the mechanism of nucleation and growth kinetics of the first-order magnetic phase transition in ordered FeRh thin film is quite similar to that of the crystallization of solids described by the Avrami model.

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Introduction

First-order phase transition has received much attentions due to not only fundamental physics but also its potential application [1–4]. It has been known for a long time that a bulk FeRh alloy exhibits a first-order magnetic phase transition between antiferromagnetic (AFM) state and ferromagnetic (FM) state upon heating or under high pressure [4]. The Curie temperature of ferromagnetic phase in FeRh alloy is around 410 °C. Because of the unique nature of such a magnetic phase transition, this alloy system has been widely studied for various physical properties for thin films and bulks. Among them, magnetic properties and structures are noteworthy mentioning. Further studies found that this transition is accompanied by a unit cell volume expansion of 1–2%, a reduction in resistivity, and a large change in entropy [4–6]. This opens up interesting possibilities for technological applications such as heat-assisted magnetic recording and microelectromechanical system devices [6–8]. However, the physical mechanism of this first-order AFM/FM phase transition in ordered FeRh-based thin films is still not very clear. To further understand the mechanism of first-order AFM/FM phase transition in ordered FeRh thin films, the magnetic properties and first-order phase transition behavior of single-crystal FeRh thin film are investigated in detail in this article.

Experimental

Single-crystal FeRh thin film with a thickness of around 100 nm was fabricated onto MgO(100) substrates using sputtering from $Fe_{50}Rh_{50}$ alloy target. The substrate temperature during deposition was kept at around 450 °C. The base pressure of the chamber was around 1×10^{-8} Torr.

The structural analysis of the films was performed by X-ray Diffractometer (XRD) using Cu Kα radiation and magnetic properties were carried out using a vibrating sample magnetometer (VSM) in fields up to 15 kOe.

Results and discussions

Figure 1a shows XRD patterns (θ–2θ) of FeRh thin film. The (001) superlattice peak and (002) fundamental peak of ordered FeRh phase are clearly observed and this result indicates that the (001)-oriented FeRh thin film was successfully fabricated on MgO (100) substrate. The other peaks with no indication in Fig. 1a come from the MgO (100) substrate. Furthermore, the ϕ-scan diffraction, as shown in Fig. 1b, confirms that the film exhibits a four-fold symmetry in the film plane, indicating that the film is of single-crystalline structure.

Figure 2 shows the temperature-dependent magnetization curves (M–T) for FeRh thin film with an applied magnetic field of 15 kOe in the direction of both perpendicular and parallel to film plane. The heating rate and cooling rate are both of 2 °C/min. Herein, We define the transition temperature, T_{tr}, as the temperature at half change of the magnetization. A sharp rise in magnetization slightly above 120 °C during heating in both directions indicates the onset of the first-order AFM to FM phase transition in FeRh single-crystal thin film. At around 140 °C, the magnetization gets saturation. The saturated magnetization is about 1370 emu/cc in perpendicular direction and during cooling the onset of the FM to AFM phase transition takes place at temperature slightly below 145 °C in perpendicular direction and 130 °C in parallel direction. It is also seen that the transition temperature (T_{tr}) in perpendicular direction is higher than that of parallel direction in both heating and cooling processes. This is most likely due to the appearance of the self demagnetization field encountered when the magnetization is perpendicular to the field which tends to retain the formation of the ferromagnetic phase. In addition, the temperature-dependent magnetization shows a hysteresis across the transition. This result confirms the first-order nature of the AFM/FM phase transition in FeRh thin film. Hysteresis across a first-order phase transition occurs due to the supercooling and/or superheating of the parent phase in the product matrix across the transition.

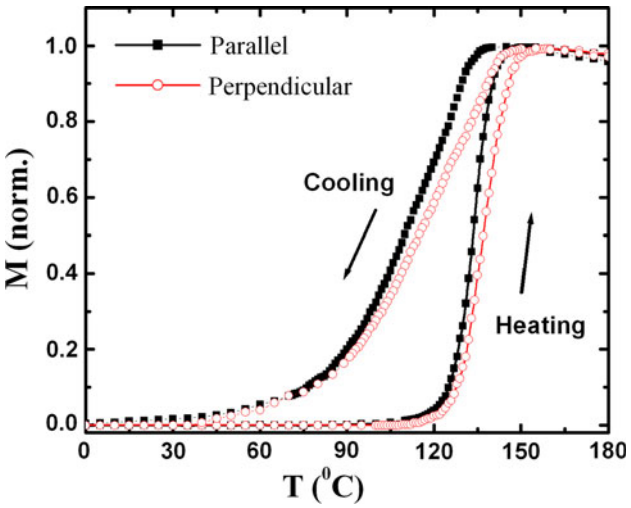

Fig. 2 Temperature-dependent magnetization curves (M–T) for FeRh thin film with an applied magnetic field of 15 kOe in the direction of both perpendicular and parallel to film plane

Figure 3 shows temperature-dependent M–H loops measured in the parallel direction of the film plane during the phase transition of FeRh thin film in both heating and cooling processes. Between each measurement the applied magnetic field is fixed to 15 kOe while temperature is changed. The hysteresis loops at the middle of transition show an obvious opening at high magnetic field which is close to saturation field. Note that there is no opening in M–H loops at the beginning of AFM–FM phase transition and the opening disappears when AFM phase has transformed to FM phase in FeRh thin film during heating. For the case of cooling, it gives similar results. Figure 4 shows

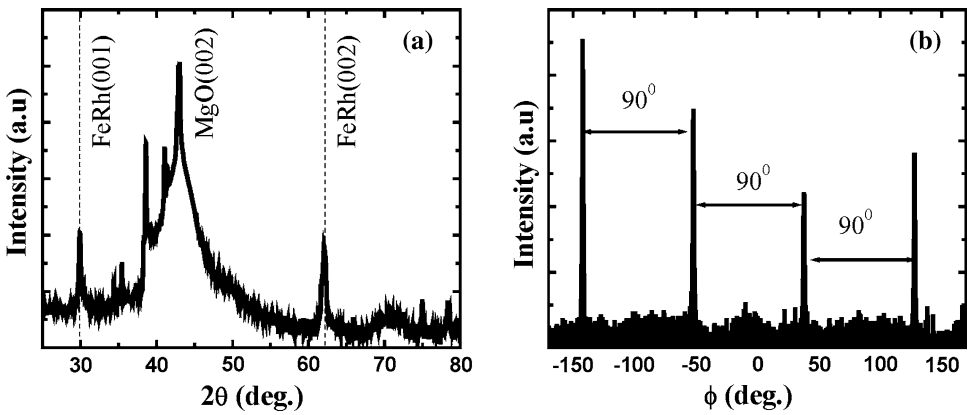

Fig. 1 XRD patterns (θ–2θ) of FeRh thin film

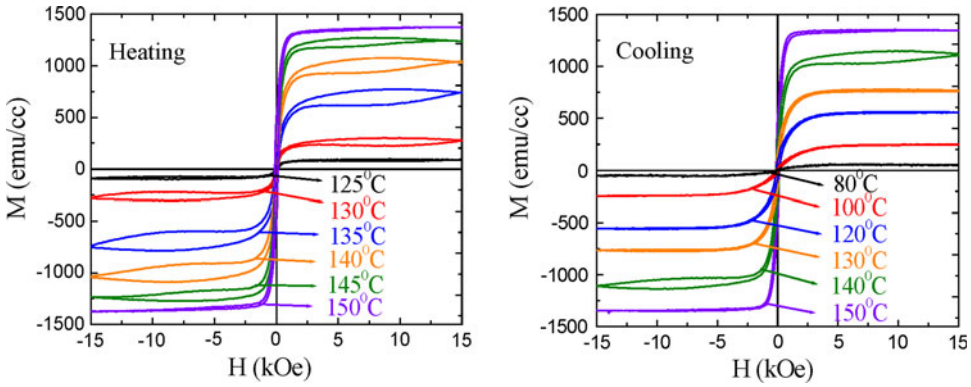

Fig. 3 Temperature-dependent *M–H* loops measured in the parallel direction of the film plane during the phase transition of FeRh thin film in both heating and cooling processes

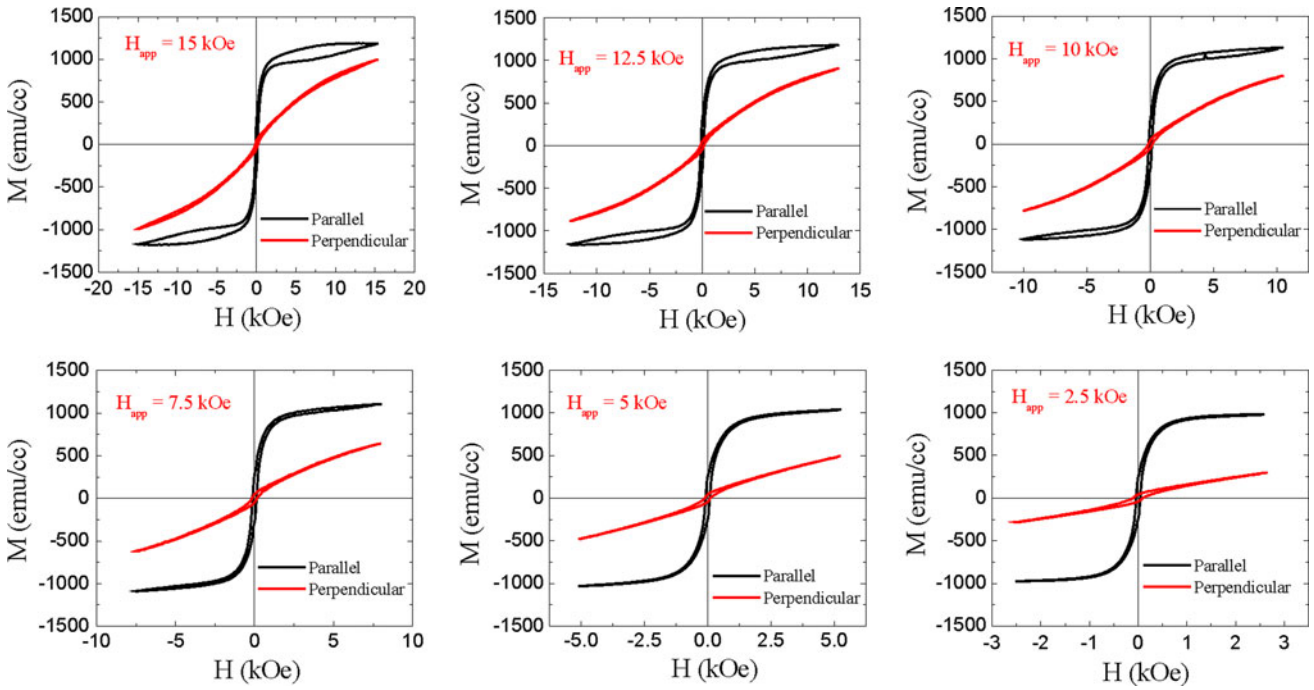

Fig. 4 *M–H* loops of FeRh thin film measured at 140 °C during heating with different applied magnetic field ranges from 2.5 to 15 kOe

the *M–H* loops of FeRh thin film measured at 140 °C during heating with different applied magnetic field ranges from 2.5 to 15 kOe. An opening in hysteresis loops can also be observed when the applied magnetic field is larger than 5 kOe. It is interesting to see that with increasing applied magnetic field from 5 to 15 kOe, the opening becomes larger. The dependence of opening in hysteresis loops on applied magnetic field and temperature indicates a reversible AFM–FM phase transition in FeRh thin film. The origin of this opening is most possible due to the magnetic-field-induced AFM–FM phase transition in FeRh thin film [5].

Having established the presence of lattice expansion and hysteresis across the transition, we focus on another signature of a first-order transition which is the existence of metastable states. The signature of metastable states can be experimentally observed by measuring the time dependence of a physical quantity intrinsic to the sample which is relevant to the phase transition. In this case, we measure the time dependence of magnetization at a fixed temperature in a fixed field using the VSM. Figure 5 shows the normalized magnetization M as a function of time during heating cycle for FeRh sample. The sample was heated up to 130 °C from a temperature well below those values where the magnetization is reversible. The magnetization was then measured as a function of time immediately after the temperature was stabilized. The normalization of M is carried out with respect to M_0, which is the magnetization value at the first measurement point after reaching 125 °C (i.e., at $t = 0$). The time-dependent magnetization follows a saturating

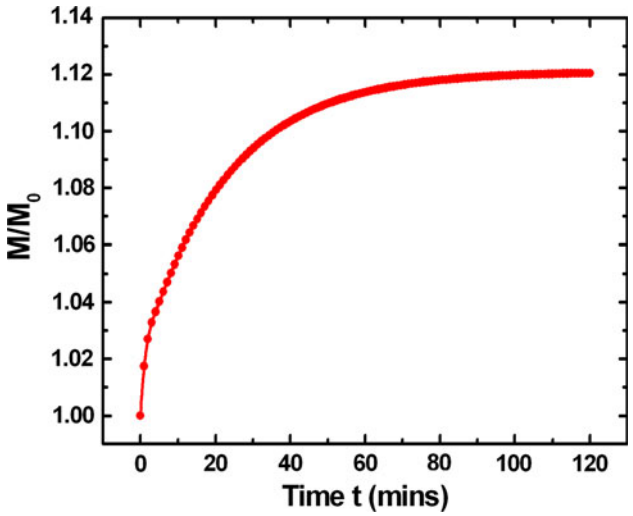

Fig. 5 Normalized magnetization M as a function of time during heating cycle for FeRh sample

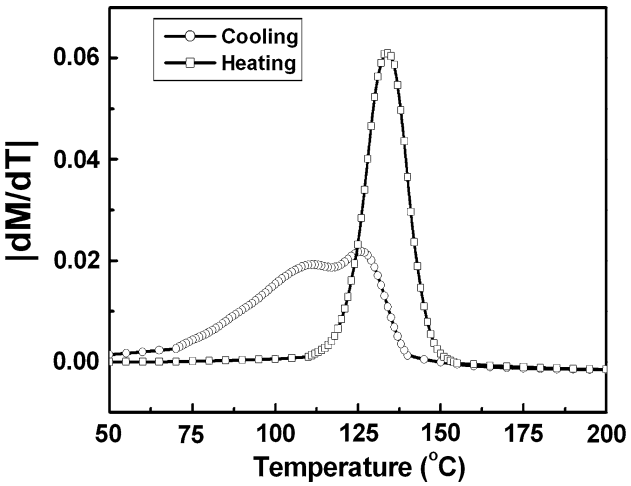

Fig. 6 Dependence of $|dM/dT|$ upon temperature for FeRh thin film

exponential law and can be very well fitted with the following equation: $M/M_0 = M_\infty - A \cdot \exp(-t/\tau)$, where M_∞ is the saturated magnetization, A is a constant, and τ is a relaxation time. The magnetization changes toward a higher value which clearly indicates the presence of a metastable (superheated) AFM phase relaxing toward the stable FM phase, and then it tends to be exhausted. This behavior could arise for the case of an AFM to FM transition when the nucleation is heterogeneous. By imaging the AFM to FM transition on a sub-micron scale in FeRh-based alloy using magnetic force microscopy [9], it has been shown the nucleation of the FM phase in the AFM matrix. Nucleation of the FM phase across the AFM to FM transition has been shown in various cases at different length scales (see [10–12]) highlighting the generality of the phenomenon. So, it can be believed that the clusters of the FM phase nucleate in the AFM matrix and vice versa during the first-order AFM–FM phase transition in ordered FeRh thin film.

Figure 6 shows the dependence of $|dM/dT|$ upon temperature in parallel direction for FeRh thin film. The data is taken from Fig. 2. Here, the value of saturated magnetization at a particular temperature was directly taken as a measure of phase fraction of the FM phase. Therefore, the nucleation and some aspects of the growth kinetics of magnetic domains during AFM/FM phase transition in FeRh thin film can be analyzed. From Fig. 6, it can be seen that during heating process, the value of $|dM/dT|$ shows an initial increase with rise in temperature and then drops with a further rise in temperature. This probably indicates that the initial part of the transition, while heating, is dominated by the creation of new FM nuclei and in the later stages these nuclei merge to grow into the product FM phase. The maximum peak in the temperature dependence of $|dM/dT|$ thus indicates the maximum growth rate of the product phase. The same discussion probably applies to the temperature dependence of $|dM/dT|$ during cooling, where the initial nucleation is dominating over the subsequent growth. The initial part of the cooling appears to be dominated by the creation of new AFM nuclei and later these AFM nuclei coalesce to form the low-temperature AFM phase (result in a rapid fall of magnetization). The results of Fig. 6 indicate a nucleation and growth mechanism quite similar to that of the crystallization of solids described in the Avrami model [13]. From the results of local imaging presented for field-induced transitions in the case of $CeFe_2$ [11]- and Gd_5Ge_4-based alloy systems [12], the nucleation and growth processes appear to closely follow the assumptions of the Avrami model. It is believed that similar nucleation and growth processes take place across the first-order phase transition in the case of FeRh thin film under consideration.

Interestingly, there is a shoulder at low temperature side during FM to AFM transition upon cooling in FeRh thin film. This feature is also observed in FeRh thin film grown on Al_2O_3 (0001) single crystalline substrate during FM to AFM transition upon cooling [5], but the origin is still not clear.

Conclusions

The magnetic properties and first-order phase transition behavior of single crystal FeRh thin film are investigated in detail in this article. The first-order magnetic phase transition is seen at a temperature of around 120 °C during heating and 145 °C cooling process in perpendicular direction. The transition temperatures in perpendicular direction is higher than that of parallel direction, which is most likely due to the arise from the self demagnetization

field encountered when the magnetization is perpendicular to the field which tends to retain the formation of the ferromagnetic phase. The *M–H* loops measured isothermally amidst the AFM–FM transition regime show an opening at high magnetic field, which indicate a reversible AFM–FM transition induced by magnetic field. The clusters of the FM phase nucleate in the AFM matrix heterogeneously and vice versa during the first-order phase transition in ordered FeRh thin film. The mechanism of nucleation and growth kinetics in the first-order magnetic phase transition of FeRh thin film is quite similar to that of the crystallization of solids described by the Avrami model.

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References

1. Zhang J, Heitmann AA, Alpay SP, Rossetti GA (2009) J Mater Sci 44:5263. doi:10.1007/s10853-009-3559-8
2. Lee CT, Zhang MS, Yin Z (2008) J Mater Sci 43:2675. doi:10.1007/s10853-007-2436-6
3. Szymczak R, Czepelak M, Kolano R, Kolano-Burian A, Krzymanska B, Szymczak H (2008) J Mater Sci 43:1734. doi:10.1007/s10853-007-2400-5
4. Kouvel JS (1966) J Appl Phys 37:1257
5. Maat S, Thiele JU, Fullerton EE (2005) Phys Rev B 72:214432
6. Lu W, Nam NT, Suzuki T (2009) J Appl Phys 105:07A904
7. Thiele JU, Maat S, Fullerton EE (2003) Appl Phys Lett 82:2859
8. Jia Z, Harrell JW, Misra RDK (2008) Appl Phys Lett 93:022504
9. Yokoyama Y, Usukura M, Yuasa S, Suzuki Y, Miyajima H, Katayama T (1998) J Magn Magn Mater 177–181:181
10. Wu W, Israel C, Hur N, Park S, Cheong S, Lozanne A (2006) Nat Mater 5:881
11. Roy SB, Perkins GK, Chattopadhyay MK, Nigam AK, Sokhey KJS, Chaddah P, Caplin AD, Cohen LF (2004) Phys Rev Lett 92:147203
12. Moore JD, Perkins GK, Bugoslavsky Y, Cohen LF, Chattopadhyay MK, Roy SB, Chaddah P, Gschneidner KA, Pecharsky VK (2006) Phys Rev B 73:144426
13. Lu W, Yan B, Huang WH (2005) J Non-Cryst Solids 351:3320