Magnetoresistance Features at the Magnetic Field-Induced Phase Transition in FeRh Thin Films

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Abstract. The causes of the appearance of first-order magnetic phase transitions remain a mystery. FeRh alloy is a classical material where a first-order magnetic phase transition occurs. The authors of this article studied the phase transition from the antiferromagnetic state to the ferromagnetic state in FeRh alloy. Comparison of the magnetometry and transport properties results allowed us to determine a number of differences in the mechanisms of the phase evolution during magnetic field and temperature induced transition. This article notes the priority of the rearrangement of the micromagnetic structure of the ferromagnetic phase as a result of the induction of a phase transition by a magnetic field. The main feature of the magnetic field induced phase transition compared to the temperature induced one is the change in the micromagnetic structure of the ferromagnetic phase. The growth of a ferromagnetic phase with less scattering fields leads to asymmetric behavior when a phase transition is induced near the metastable state. We also focused on the importance of taking into account the effect of magnetostriction when analyzing the evolution of the phase transition, which leads to the irreversibility of the phase transition near a zero magnetic field.

Keywords: *first-order phase transition; magnetoresistance; thin film; iron-rodium; magnetization.*

1 Introduction

Metals or compounds with a magnetic first-order phase transition exhibit an anomalous change in caloric, magnetic, and structural properties near the phase transformation temperature. The temperature at which a sharp change in physical properties is observed can be changed by varying the elemental composition, the stoichiometry of the crystal lattice, the external stresses, and the external magnetic or electric field. Therefore, such materials, which can be called functional, are practically significant in the field of energy (as elements for solid-state cooling systems, thermomagnetic generators) [1,2], sensors [3] (temperature and pressure sensors), microelectronics (hard disk elements in

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heat-assisted magnetic recording (HAMR) technology) [4] and medicine (targeted drug delivery, hyperthermia, hypothermia) [5,6].

The well-known $Fe_{49}Rh_{51}$ alloy was selected as the object of this investigation because it exhibits a negative giant magnetocaloric effect, which change the temperature of the sample down to -7 K to -8 K [7] in an external magnetic field of up to 1.95 T near 300 K, the symmetry of the crystal structure does not change during the first-order phase transformation, and such an alloy has a binary elemental composition. These features allowed us to build a simple first-order magnetic phase transition descriptive model. The FeRh compound has been investigated for a long time and its stationary properties are well reported [7]. Of great interest are the processes that are observed near the temperature of the magnetic phase transition from the antiferromagnetic state to the ferromagnetic state. Previously, it has been established that this transition to the ferromagnetic state is accompanied by the processes of nucleation, growth, and phase merging [4,8,9]. However, the features of the ferromagnetic phase evolution during the phase transition are still poorly understood. For example, in the aforementioned papers, attention is paid to the study of a temperature-induced phase transition. The articles [4,9] present the results of the phase transition behavior in the metastable state with a slight change in the external magnetic field (less than 1 kOe). Therefore, the question of possible differences in the magnetic phase transition realized only by a high external magnetic field remains open.

In the literature, the question of the phase transition evolution in iron-rodium alloys has mainly been considered in thin films. For example, it was concluded in [10] that the magneto-structural phase transition does not occur in one step but as a result of a multistage process in which various mechanisms of the nucleation and growth of a new ferromagnetic phase manifest themselves. The evolution of the antiferromagnetic–ferromagnetic transition of FeRh film was studied by the authors of [11] using different X-ray techniques (Xray-PEEM, XMCD, nano-c methods). They established that, during the antiferromagnetic–ferromagnetic magnetostructural transition, ferromagnetic clusters are nucleated on defects and its further increase is delayed by the surrounding region in which the phase transition has not yet occurred. We note that this process leads to a wide region of phase coexistence during the transition as well as to a temperature dependence of the magnetic domain size, almost the same as that observed in second-order phase transitions. The change in the phase transition temperature under the influence of external mechanical stresses was demonstrated in [12]. It is also worth noting that a number of interesting works [13-17], where the result of the magnetoresistance measurements for FeRh alloys, are presented. The phase transformation from the antiferromagnetic state to the ferromagnetic state is accompanied by an increase

in conductivity. Measurements of transport properties are a powerful tool for studying the features of phase transitions and the magnetic structure in certain objects [18,19].

Significant differences in the behavior of thin-film and bulk FeRh alloy magnetoresistance were first discovered in 1966 [20]. Subsequently, interest in studying the transport properties of thin films grew substantially. The issue of the influence of high magnetic fields (up to 37 T) on the conductivity of FeRh compounds was studied [21]. Later, relations between the caloric, transport, and magnetic properties of such compounds were revealed, and the parameters of the elastocaloric effect were determined [22]. The correspondence between magnetization and resistivity in thin films was demonstrated in [23]. A distinctive feature in the transport properties during phase transition in thin films is the presence of mechanical stresses from the substrate [24]. The degree of the residual stresses can be changed by annealing [25] or by choosing the substrate's crystal structure [26]. At present, notable attention is paid to the study of films on ferroelectric substrates [27-29]. Such functional samples may be used as elements for topological antiferromagnetic spintronics [15,30]. A mathematical model that describes the transport properties in iron-rhodium alloys is described in [14]. It uses a model that takes into account magnetic moments fluctuation and thermal lattice vibrations. However, the predictive power of such a model is limited, since the phase transition in this alloy is sensitive to its microstructure. It follows that the electrical properties of FeRh alloy are an important aspect that must be taken into account when studying phase transitions in such systems.

In this study, an attempt was made to experimentally establish the magnetoresistivity features of external magnetic field induced antiferromagnetic-ferromagnetic phase transformation in a thin film of FeRh alloy.

2 Experiments

A thin FeRh film (56 nm thick) was studied in this work. The film was deposited by the molecular epitaxy method. The film was grown on an MgO (001) single-crystal substrate. The substrate was annealed at a temperature of 873 K for 1 hour before the deposition process. The alloy was deposited by co-evaporation of Rh and Fe on the substrate. The substrate temperature was 773 K, in a vacuum chamber. The pressure in the chamber was kept at ~10^{-10} Torr. The film was annealed at a temperature of 893 K for 1.5 hours after synthesis. A detailed structural analysis can be found in [4]. The XRD results confirmed the presence of a CsCl type crystal lattice. Additional results from EDX analysis refined the elemental composition of the alloy as $Fe_{49}Rh_{51}$. It is also worth

noting that the presence of a paramagnetic gamma phase was not detected according to the results of the structural analysis. Previously, it has been shown that the gamma phase of iron-rhodium is in a paramagnetic state [31].

The temperature/field dependencies of the magnetization were obtained using a LakeShore 7407 Series VSM. Detailed measurement protocols can be found in [4]. Transport properties were measured with PPMS (Quantum Design). The sweeping rate of the magnetic field was 25 Oe/s. The measurements were made according to two protocols (measurements during heating and measurements during cooling). According to the first protocol, the sample was heated from a temperature of 300 K in the zero external magnetic field (completely antiferromagnetic state) to a temperature T, at which the magnetoresistance was measured. The sample was cooled to its original state in the zero external magnetic field after each measurement. According to the cooling protocol, the sample was heated without magnetic field to a temperature of 385 K (fully ferromagnetic state) and then cooled to a temperature T, at which the magnetoresistance was measured. The sample was heated to its initial state in the zero external magnetic field after each measurement.

3 Results and Discussion

As noted above, the structural characterization results are presented in [4]. It was determined there was a CsCl-type crystal lattice in the sample. An elemental analysis was also carried out [4]. The composition of the alloy was $Fe_{49}Rh_{51}$ according to the results of the EDX analysis. Additionally, no additional phases were found using this method.

The magnetization temperature dependence (in the range from 300 K to 430 K in the external magnetic field of 1 kOe and 16 kOe) is shown in Figure 1. The alloy showed antiferromagnetic behavior below 320 K. A ferromagnetic state of the alloy was observed at temperatures above 380 K. The average temperature of the phase transformation from the antiferromagnetic to the ferromagnetic state was 368 K in field 1 kOe. The average temperature of the phase transition from the ferromagnetic state to the antiferromagnetic state was 342 K. Thus, the temperature hysteresis is 24 K. As can be seen from Figure 1, the presence of a temperature hysteresis and the value of the phase transition temperature, which depends on the external magnetic field, indicate a first-order phase transformation. The coexistence of antiferromagnetic and ferromagnetic phases was seen in this alloy near the temperature of the phase transition. The boundary transformation between the phases should provide conductivity changes. We measured the magnetic field dependencies of the resistivity at different temperatures to establish the main features of the magnetic phase transformation

evolution. We calculated the resistivity from the resistance, knowing the geometry of the sample and the distance between the contacts.

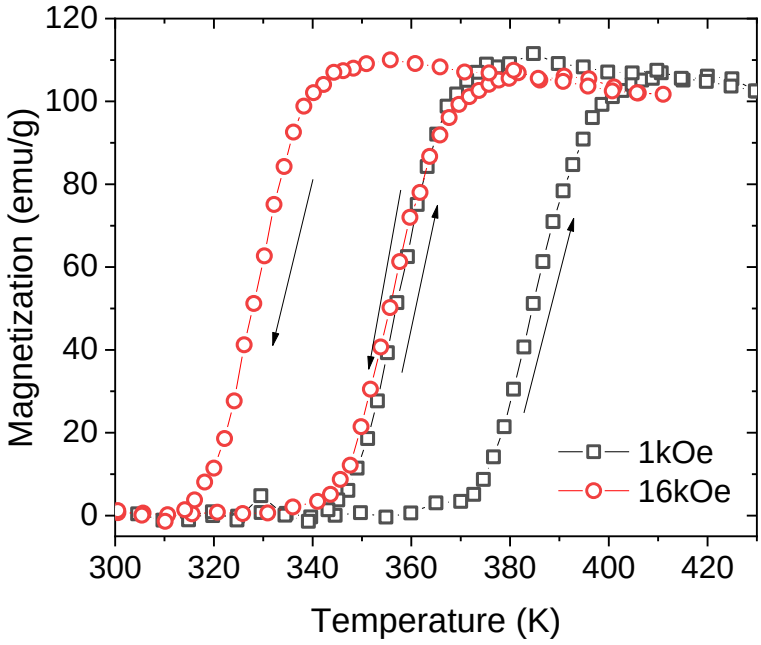


Figure 1 Temperature dependence of magnetization in the external magnetic fields of 1 kOe and 16 kOe.

It was demonstrated in [4] that ferromagnetic clusters appeared on the film surface during cooling occurs at a temperature of 350 K. Therefore, we started the measurements of the magnetoresistivity (field dependence of the resistivity) at this temperature (Figure 2). At the beginning of the measurements, an increase in resistivity was observed, which may be described by a transform in the micromagnetic state of the ferromagnetic phase. We assumed that the maximum of the resistivity is reached when the number of magnetic domains in the ferromagnetic phase becomes maximal. A further increase in the external magnetic field (from ~0 kOe to 90 kOe) leads to the transition of the alloy to a fully ferromagnetic state, which is accompanied by a decrease in magnetoresistivity. Also, we note that as the external magnetic field decreases (from 90 kOe to 0 kOe), the magnetoresistivity curves (Curve 1 and Curve 2 in Figure 2) do not coincide. This may indicate a change in the mechanisms of the evolution of the phase transition during application and removal of the magnetic

field. Residual mechanical stresses occur due to the magnetostriction effect [22] (in addition to the volumetric expansion of the crystal lattice during heating) as a result of inducing the magnetic phase transition by the magnetic field. These conclusions are also valid for thin films according to the structural studies presented in [24,32]. The phase transition temperature increases in regions where mechanical stress is present [12]. This leads to the fact that the antiferromagnetic phase occurs in lower fields (in Curve 2 at Figure 2). Curves 1 and 3 in Figure 2 are symmetrical. We decided to analyze in detail the field dependencies of the resistivity during heating since there is an asymmetry in the phase transformation evolution under the transition from the antiferromagnetic to the ferromagnetic state, and vice versa [33].

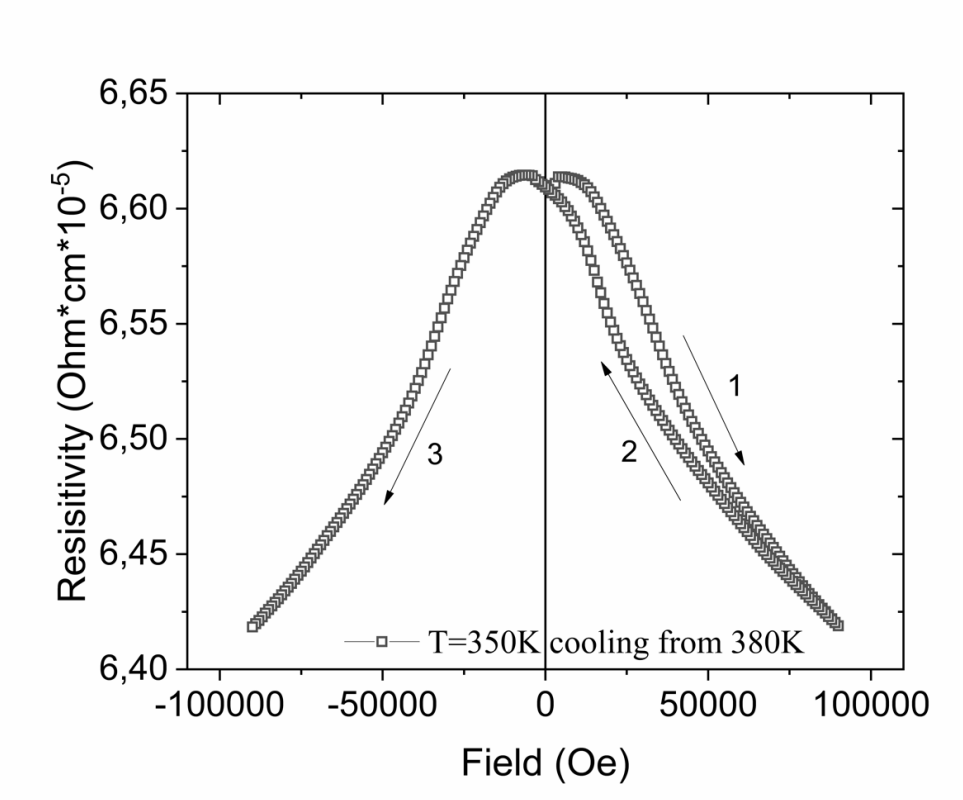
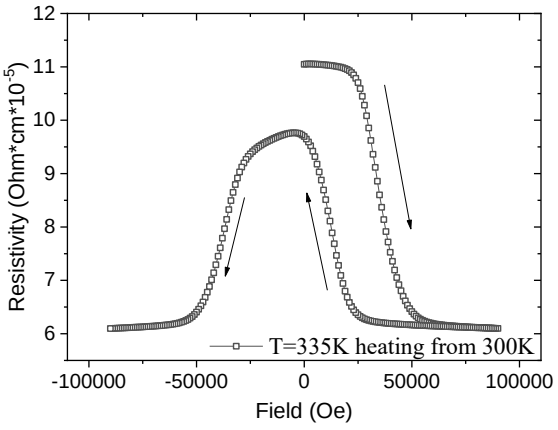


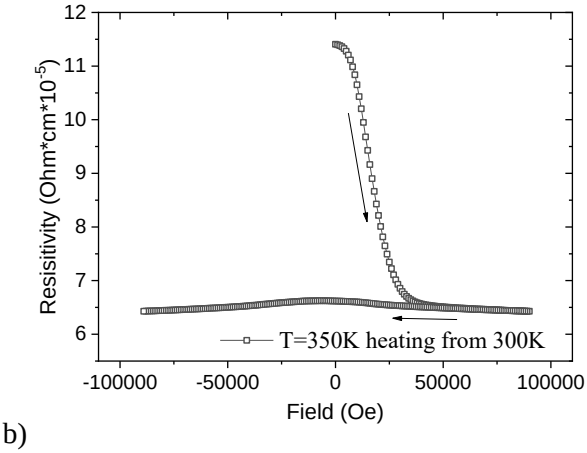
Figure 2 Field dependence of resistivity at a temperature of 350 K.

Figures 3a-c show the dependencies of the magnetoresistivity at temperatures of 335 K (AFM state), 345 K, 350 K (measured during heating). The initial state (in zero external magnetic field) was highly resistive for all temperatures (Figure 3). The large value of the resistivity is associated with the fact that the antiferromagnetic phase and the ferromagnetic phase coexist in this state. An

increase of the magnetic field leads to the appearance of a phase transition and a decrease in resistivity. The irreversible change of the resistivity value when the magnetic field decreases from maximum to zero is due to the presence of a temperature hysteresis of the phase transition. As the magnetic field decreases, the volume of the antiferromagnetic phase does not reach its initial value. A regular trend was also observed, according to which the resistivity in the zero external magnetic field (after returning from 90 kOe) decreased with increasing temperature. Also, it should be noted that the asymmetry of the curves in the external magnetic field ranged from 20 kOe to -20 kOe. The sharp increase in the resistivity in the range from 20 kOe to 0 kOe may be associated with an increase in the volume fraction of the antiferromagnetic phase. The further decrease in resistivity in the external magnetic field from 0 kOe to -20 kOe may be associated with the processes of ferromagnetic phase nucleation and changes in its micromagnetic structure, which is accompanied by a slight decrease in resistivity. We focused on the behavior of the resistivity in this range of fields, which are presented in Figure 2 and Figure 3a. The change in the micromagnetic structure occurred at fields below 1 kOe when the film was re-magnetized in the ferromagnetic state (Figure 2). It is necessary to apply the external magnetic field 20 kOe to orient the magnetic moments in ferromagnetic clusters when the film obtains a predominantly antiferromagnetic state (Figure 3a). Such significant differences in the phase transition evolution may be associated with the presence of unidirectional magnetic anisotropy between the antiferromagnetic and ferromagnetic phases in the film [33]. Visualization of the differences between the evolution of the phase transition during cooling and heating in low magnetic fields is presented in [34] using Kerr and MFM microscopy.



a)



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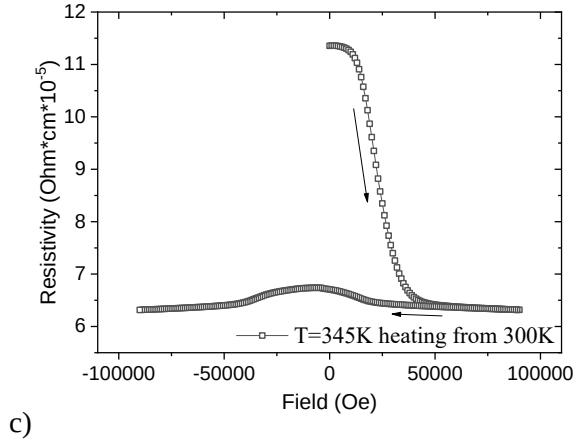

c)

Figure 3 Field dependencies of resistivity at temperatures of a) 335 K, b) 345 K, c) 350 K.

An increase in the external magnetic field from -20 kOe to -50 kOe was accompanied by a sharp decrease in resistivity (Figure 3a). This fact may be described by the presence of ferromagnetic phase growth processes. These results are in good agreement with the model of evolution of the ferromagnetic phase described in [4,35].

It should be noted that the main features in the magnetoresistivity dependencies for thin-film samples have not been observed in bulk alloys previously [21,35]. First, in thin films, the presence of a residual ferromagnetic phase is clearly pronounced when the external magnetic field is removed. This can be seen in Figure 3. The resistivity values near the zero field do not match when the magnetic field is switched. As noted above, this can be explained by the presence of mechanical stresses from the substrate in the film sample. Secondly,

the relative change in resistivity in the film sample (77%) is smaller than in the bulk alloy (~85%) [35]. There are several reasons that may explain this result. A thin film has a more perfect crystal structure than a bulk alloy (as a rule, bulk FeRh alloys contain additional paramagnetic crystallographic phases [36]). An important role is also played by the residual antiferromagnetic phase near the substrate [4]. Its presence prevents a decrease in the magnetoresistivity. The revealed features of transport properties in FeRh thin films should be taken into account in further possible practical applications in spintronics.

4 Summary

Analysis of the field dependencies of the resistivity allowed us to clarify a feature of the phase transition evolution. The field-induced phase transformation had several differences compared to the thermally assisted one. The presence of a magnetic field causes the appearance of an additional stage in the evolution of the ferromagnetic phase. For example, there is a rearrangement of the ferromagnetic phase domain structure. Also, the field-induced phase transition demonstrates irreversibility due to the presence of magnetostriction effect. The significant asymmetry in the magnetic phase transition evolution during heating and cooling can be explained by the fact that the change in the ferromagnetic phase micromagnetic structure is determined by the value of the unidirectional magnetic anisotropy in the metastable state. The obtained results also confirmed the validity of the previously described model of the ferromagnetic phase evolution in thin films. The revealed differences in the magnetotransport properties of thin films and bulk samples will make it possible to select more optimal modes of operation of FeRh-based spintronic elements.

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transport properties, validation; **N.S.P.** performed the formal analysis, and provided supervision.

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