Depth Profile of the Phase Transition of the FeRh Alloy in FeRh/BaTiO$_3$

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Abstract

We report on the depth dependence and technological limits of the phase transition of the iron rhodium alloy as function of temperature, external magnetic and electric fields in the FeRh/BaTiO$_3$ multiferroic, determined by grazing-incidence nuclear resonant scattering measurements. The change of temperature induces a continuous and homogenous antiferromagnetic / ferromagnetic phase transition through the entire FeRh layer, except in the near substrate region. External magnetic field does not affect this mechanism, but the application of electric field changes it fundamentally: the phase transition of the alloy suddenly propagates from the substrate up to a height, defined by the combination of temperature and external magnetic field, as soon as the applied electric field reaches $\sim 20$ kV/m.

**Keywords:** FeRh alloy, depth dependent phase transition, depth structure, nuclear resonance scattering, multiferroic

1. Introduction

By 2030, the global electricity consumption is estimated to increase by $5 \times 10^3$ TWh [1, 2]. Ignoring such a trend could have a serious impact on the environment, but this dramatic increase could be curbed by the use of new technologies and materials, that help to optimize the performance of electronic devices [3]. Composite multiferroics, which are composed of two materials of different ferroic properties [4, 5], are among the novel materials that can be used to increase the efficiency of devices based on the principles of spintronics [6, 7, 8], magnetic switching [9, 10, 11], magnetic refrigeration [12, 13, 14, 15], biomechanical energy harvesting [16, 17, 18] and photovoltaics [19, 20, 21].

The iron rhodium alloy is one of the most promising component of the composite multiferroics, due to its technologically exploitable mechanical and magnetic properties [22]. The equiatomic FeRh alloy has a temperature induced, fully reversible, antiferromagnetic (AFM) $\leftrightarrow$ ferromagnetic (FM) magnetic phase transition in the transistor-based devices operating temperature range [23, 24, 25]. At room temperature the (equiatomic) FeRh is in AFM phase with
a CsCl-type bcc-based B2 crystal structure [26]. As the alloy is heated up, the volume ratio of the FM phase increases gradually until the fully ferromagnetic state is reached around 383 K; and vice versa, as the alloy cools down, the volume of the AFM phase increases. Between 299 K and 393 K, there is favored AFM / FM phase ratio for each temperature, therefore the change of temperature triggers a partial phase transition. In addition to the changing magnetic order, the transition of the FeRh has effects on the other properties of the alloy as well. First: it involves the changing of the magnetic moments of iron and rhodium atoms from the room temperature AFM values of 3.2 µB (Fe) and 0.0 µB (Rh), to the high temperature FM values of 3.1 µB (Fe) and 0.9 µB (Rh) [27]. Second: the phase transition directly influences the electrical resistivity of the FeRh [28]. Third: the FM → AFM phase transition is accompanied by the decrease of lattice parameter by about 1% [29, 30]. By reversing the third phenomenon, mechanical compression can be used to trigger the FM → AFM magnetic phase transition of the alloy [31, 32]. To create a multiferroic composite, the FeRh alloy is often coupled with the piezoelectric BaTiO₃ (BTO) ceramic [22, 33], whose epitaxial coupling allows the alternation of the phase composition of the alloy (along with its above listed consequences) with external electric field via piezoelectric strain [22, 34, 35, 36].

Notwithstanding its exceptional properties, the optimal utilization of the FeRh/BTO multiferroic requires the in-depth knowledge of how the phase composition of the FeRh alloy changes as a function of technologically important external parameters (temperature, magnetic and electric fields). The phase transition of the alloy on its surface could be efficiently imaged by either magnetic force [37] or X-ray photoemission electron microscopy [38], so the horizontal map of its AFM / FM structure is now thoroughly explored [39, 40]. In contrast, only a handful studies [41, 42, 43, 44] describing the depth dependent AFM / FM phase structure of the FeRh layer are available, while no studies were devoted to investigate the depth profile of the phase transition as a function of temperature, external magnetic and electric fields. The best description of the phase selective depth profile of the FeRh was given by a polarized neutron reflectivity (PNR) study [43], according to which the magnetic structure of the alloy in the FeRh/MgO composite is vertically inhomogeneous at room temperature, with a thin FM layer near the substrate. This FM phase may be an effect of the epitaxial strain caused by the lattice mismatch between the substrate and the FeRh layer [45, 46, 47], so it could be suspected that the depth profile described in the PNR study varies depending on the substrate used. Therefore, for the optimal utilization of the FeRh/BTO composite multiferroic, a comprehensive study was still needed to reveal the depth profile of the phase transition of the alloy as a function of technologically important external parameters.

In this work, the FeRh layer was deposited by molecular beam epitaxy (MBE) on the BTO substrate; and its atomic composition was determined by Rutherford backscattering spectrometry (RBS). The overall crystal structure and lattice parameters of the alloy were determined by X-ray diffractometry (XRD), while the thickness of the film was determined by X-ray reflectivity (XRR). The magnetic properties of the alloy were determined by vibrating sample magnetometry (VSM); and the iron microenvironments were determined by conversion electron Mössbauer spectroscopy.
Finally, the depth profile of the phase transition of the FeRh was investigated by grazing-incidence nuclear resonance scattering (GI-NRS).

2. Experimental

2.1. Sample preparation

The BaTiO$_3$(100) substrate (5 mm $\times$ 5 mm $\times$ 1 mm rectangular) was purchased from Alineason – Material & Technology. Before the sample preparation, the substrate was cleaned in ultrasonic ethanol bath, then it was baked out under ultra-high vacuum conditions at 873 K for 1800 seconds. The $^{57}$FeRh alloy of 18.0 nm nominal thickness was deposited on the BTO substrate with the MBE apparatus of the Wigner Research Centre for Physics (Wigner RCP). To achieve better lateral homogeneity, the substrate was continuously rotated. Electron gun for $^{103}$Rh, and effusion cell for $^{57}$Fe evaporation were used, at growth rates of 0.05 nm/s for both metals. During deposition the temperature of the substrate was kept at 903 K, while the pressure in the growing chamber never exceeded $3.8 \times 10^{-9}$ mbar. The quality of the epitaxial growth was monitored by in-situ reflection high-energy electron diffractometry (RHEED). Later a 20 nm thick gold layer was deposited on the opposite side of the BTO substrate for electric contact.

2.2. Characterization methods

2.2.1. Rutherford backscattering spectrometry measurement

The RBS measurement of the $^{57}$FeRh layer was performed by using 2 MeV $^4$He$^+$ ion beam obtained from the 5 MV Van de Graaff accelerator of the Wigner RCP. The beam was collimated to the necessary dimensions of 0.5 $\times$ 0.5 mm$^2$ with two sets of four-sector slits and the measurements were performed with an ORTEC detector of a solid angle of 4.657 msr. The dose of the measurement was 20 µC. The ion current of typically 8 nA was measured by a transmission Faraday cup [48]. To reduce the surface contamination, liquid N$_2$ trap was used. The pressure in the scattering chamber was about $1.5 \times 10^{-6}$ mbar during the experiments. The RBS data were evaluated by the RBX code [49].

2.2.2. X-ray diffractometry measurement

XRD experiment was carried out in the Centre for Energy Research using a D8 Discover (Bruker AXS, Karlsruhe, Germany) diffractometer. Cu Kα radiation ($\lambda = 1.5418$ Å) was used for the measurement. To decrease beam divergence and to improve the parallelism of the beam, 1 mm slits were used at the source and the detector. Furthermore, a 90° rotated Soller slit was also installed between the sample and the detector-side slit. At last, a secondary monochromator was used at the detector side, to achieve better signal-to-noise ratio. For the evaluation of the XRD results, the Diffrac.EVA [50] program was used.

2.2.3. Conversion electron Mössbauer spectrometry measurement

CEMS measurement was performed by using a conventional WissEl/DMSPCA Mössbauer spectrometer, operated in sinusoidal drive mode at drive frequency 16 Hz, at the Wigner RCP. The
activity of the $^{57}$Co(Rh) single-line Mössbauer source was around 925 MBq at the time of the measurement. The resonant conversion electrons were detected with a custom-built gas-flow single-wire proportional counter of 1 mm distance between sample and anode wire, working with a mixture of 96 % v/v He and 4 % v/v CH$_4$ gas at bias voltage 924 V. The distance between the source and the sample was 53 mm. The spectrum was evaluated by the MossWinn 4 code [51].

2.2.4. Grazing-incidence nuclear resonance scattering and X-ray reflectometry measurements

GI-NRS and XRR experiments were carried out at the Nuclear Resonance beamline [52] ID18 of the European Synchrotron Radiation Facility (ESRF). The measurements were performed in 4 bunch mode at 14.414 keV, the energy corresponding to the 1/2 ↔ 3/2 nuclear transition of $^{57}$Fe, with a beam of 0.5 meV energy bandpass. The beam was focused by a Kirkpatrick-Baez mirror system both horizontally and vertically to 20 μm and 8.7 μm, respectively. The sample was mounted in a custom-built vacuum chamber (Fig. 1) that allowed temperature (273 K – 400 K) control and adjustable electric (between 0 V – 200 V) and magnetic (0 mT – 150 mT) fields to be applied on the sample during the measurements. The GI-NRS quantum-beat patterns and XRR reflectograms were analyzed by the in-house (Wigner RCP) developed FitSuite program [53].

![Fig. 1. Schematic of the sample holder designed for the GI-NRS experiment](image)

The GI-NRS experiment was performed in four groups of measurements. The 1$^{st}$ group was performed without applying any external electric or magnetic field. The temperature of the sample was raised directly to 394 K, then it was gradually lowered to 299 K, meanwhile GI-NRS quantum-beat patterns were recorded at three different grazing angles (3.49 mrad, 4.19 mrad and 4.71 mrad), at each temperature step (cooling phase). After this, the temperature of the sample was gradually raised back to 394 K, again with 3 – 3 GI-NRS measurements, at each selected grazing angle, at each temperature step (heating phase). The 2$^{nd}$ group of GI-NRS measurements were performed similarly to the 1$^{st}$ ones, but with a 150 mT external magnetic field applied to the sample. During the 3$^{rd}$ group of GI-NRS measurements, the sample was heated up to 394 K; then the voltage in
the electric contacts (yellow parts on Fig. 1.) was gradually increased up to 100 V (i.e. 0 - 100 kV/m electric field ($E$) was generated on the 1 mm thick BTO substrate), meanwhile 3 – 3 GI-NRS measurements were performed, at each selected grazing angles, at each voltage step. After the 100 kV/m measurements, the voltage was switched off, and the sample was cooled down to the next temperature step - then the whole electric field build up process was repeated again. At last, the 4\textsuperscript{th} group of GI-NRS experiments were performed in the same way as the 3\textsuperscript{rd} ones, but with 150 mT external magnetic field applied.

2.2.5. \textit{Vibrating sample magnetometry measurements}

The magnetic moments were measured with a vibrating sample magnetometer from Quantum Design in a Physical Properties Measurement System equipped with a 9 Tesla magnet (PPMS-9T) at Complutense University of Madrid (UCM). All VSM measurements were performed in five orientations: four times in in-plane orientations [(100), (010), (110) and (-110)] and once in perpendicular direction (001), in the order listed. Magnetic hysteresis loops were measured in 10 K steps in the temperature range from 300 K to 400 K.

3. \textbf{Results & discussion}

3.1. \textit{Sample characterization}

The RHEED diffractogram of the $^{57}$FeRh/BaTiO$_3$(100) composite shows a linear structure characteristic of epitaxial layers (“A” on Fig. 2.), which confirms high level epitaxy between the alloy and the ceramic.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig2.png}
\caption{“A” RHEED diffractogram; “B” $^4$He$^+$ RBS spectra at tilt angle of 7°, and “C” XRD pattern of the $^{57}$FeRh(B2)/BaTiO$_3$(100) composite}
\end{figure}

The actual thickness of the $^{57}$FeRh layer and the atomic ratio of the alloy was determined by RBS (“B” on Fig. 2.) and was found to be 15.7 ± 0.3 nm and $^{57}$Fe$^{0.508 \pm 0.023}$Rh$^{0.492 \pm 0.022}$, respectively.

The XRD pattern shows the B2 type FeRh(001) peak around 30°, the FeRh(002) peak around 61 and the BaTiO$_3$ (200) peak around 45° (“C” on Fig. 2.). The lattice parameter was estimated to be 3.0361 ± 0.0004 Å, which is close to the value reported for the B2 FeRh phase [54]. Since no peak corresponding to other possible structure of FeRh (i.e.: to the paramagnetic A1 phase) was
found in the pattern, our alloy can be considered structurally homogeneous. Together, RHEED, RBS and XRD results confirmed the successful, high-quality sample preparation.

3.2. Determination of the (sub)layer structure of the FeRh/BTO composite

For the GI-NRS model, the iron microenvironments of the alloy were determined by CEMS. Based on previous studies [40, 55, 56] and the RBS results, four binomial distributions, one singlet, one sextet, and a doublet were used for evaluation of the 299 K CEMS spectrum (“A” on Fig. 3.). The Mössbauer parameters [table 1. in supplementary materials] of the first six spectral components are characteristic to the ferromagnetic and antiferromagnetic iron rhodium phases [55, 57], while the parameters of the doublet indicate the presence of a separate paramagnetic oxide phase. The unusual existence of the FM phase at room temperature, which is common in the case of thin FeRh films, caused by the small lattice mismatch between the BTO and the FeRh, as it was previously reported in [41, 43, 58, 59]. The detailed description of the model used for the deconvolution of the CEMS spectrum can be found in the supplementary materials.

![Image](https://example.com/image.jpg)

**Fig. 3.** 299 K CEMS spectrum “A”; and X-ray reflectogram “B” of the sample; and the arrangement of the (sub)layers in the GI-NRS model “C”, the number after the sublayer shows the depth of its bottom within the whole FeRh layer.

The analyses of the XRR reflectogram (“B” on Fig. 3.) revealed that the actual layer structure of the sample is Fe$_3$Rh$_9$O$_{2x}$[(0.7 ± 0.1) nm]/$^{57}$Fe$_{0.3}$Rh$_{0.7}$[(16.8 ± 0.1) nm]/BaTiO$_3$(100), where “Fe$_3$Rh$_9$O$_{2x}$” is an unspecified iron–rhodium oxide. It is noticeable, that the thickness of the alloy layer is different in the RBS and XRR results. This difference can be explained by the fact that the RBS evaluation used the density of the bulk FeRh to calculate the layer thickness, whereas the density may differ slightly for thin films. Based on this, for the model used for the analysis of the GI-NRS quantum-beat patterns (illustrated as “C” on Fig. 3.) the layer thickness values were taken from the XRR results, not from those determined by RBS.

In the GI-NRS model, to be able to model the depth profile of the phase transition, the FeRh layer was divided into ten sublayers, each representing the 1/10 of its thickness (“C” on Fig. 3.). Each of these sublayers is described as a homogeneous composition of all (spectral)components from CEMS results (excluding the oxide component). During the analysis of the GI-NRS quantum-beat patterns, the Mössbauer parameters of the corresponding components were kept at the same
values in each sublayer, taking into account the temperature-dependent changes of the parameters. In addition, within the FM and AFM phases, the ratios of the microenvironments were fixed in each sublayer at the ratios that were derived from CEMS. As a result, the only two fitting parameters which could differ between the individual FeRh sublayers (at any given temperature/electric/magnetic field combination) were the relative amounts of the FM ($X_{\text{FM}}$) and AFM iron rhodium phases ($X_{\text{AFM}}$). As $X_{\text{FM}}$ was modeled as $1-X_{\text{AFM}}$ in each sublayer, the only remaining independent parameters were $X_{\text{AFM}}$ (layer), which is used in this article as a single parameter to describe the phase composition of the sublayers.

By simultaneously evaluating the 3–3 corresponding GI-NRS quantum-beat patterns; that were measured with the same experimental parameters, but at different grazing angle; $X_{\text{AFM}}$ values were determined individually in each FeRh sublayer, at each temperature, based on similar principles that were described in [60, 61, 62]. In Fig. 4, we can see the calculated intensity (given in arbitrary units) in depth of the sample as the function of energy detuning from the resonance (Mössbauer drive velocity). It is apparent that at the resonance peaks the penetration is quite shallow, but we have to keep in mind, that in the measured quantum beat patterns the whole broadened lines have role and not only their peaks.

![Fig. 4. Calculated information depths of the GI-NRS measurement at grazing angles of 3.49 mrad “A”, 4.19 mrad “B” and 4.71 mrad “B”](image)

Each calculated $X_{\text{AFM}}$ values, along with their uncertainties, in each temperature (and magnetic/electric field combination), can be found in tables 3.–16. of supplementary materials. The fitting of our model to the measured data points, as well as the effects of the different grazing angles, external magnetic and electric fields on the GI-NRS quantum-beat patterns are demonstrated on Fig. 5.
Fig. 5. The effect of the different grazing angles and external magnetic/electric fields on the GI-NRS quantum-beat patterns; during the cooling phase at 352 K; dots as measured and lines as modelled; “A” at 0 mT applied external magnetic field and 0 kV/m applied electric field at 4.19 mrad grazing angle; “B” at 0 mT, 0 kV/m, 3.49 mrad; “C” at 0 mT, 0 kV/m, 4.71 mrad; “D” at 0 mT, 100 kV/m, 4.19 mrad; “E” at 0 mT, 100 kV/m, 3.49 mrad; “F” at 0 mT, 100 kV/m, 4.71 mrad; “G” at 150 mT, 0 kV/m, 4.19 mrad; “H” at 150 mT, 0 kV/m, 3.49 mrad; and “I” at 150 mT, 0 kV/m, 4.71 mrad

3.3. Depth dependent determination of the phase transition of the alloy as a function of temperature, without external magnetic or electric field

The determined $X_{\text{AFM}}$ per layer values show, that the 1st and the 10th FeRh sublayers contain very little AFM phase, regardless of whether this phase overwhelmingly dominates the $^{57}\text{FeRh}$ alloy at 299 K without applied magnetic- or electric fields (dark blue bars on Fig. 6). This observation corresponds well to the previous PNR study, where similar depth structure was described in comparably thick FeRh films [43].
The analysis of the further GI-NRS quantum-beat patterns has also revealed, that these inherently low $X_{\text{AFM}}$ values observed in the 1st and 10th sublayers barely change regardless of temperature of the measurement (Fig. 6). This result indicates that the magnetic structures of these sublayers, at bottom and top of the FeRh layer, are stabilized in FM state due to interface related effects. In the case of the 10th sublayer, this effect may be explained by the small lattice mismatch between FeRh and BTO inducing an epitaxial strain which favors the FM state, by the same mechanism as described in [63, 64, 65, 66]. Meanwhile, in the case of the 1st sublayer, oxidation based processes may be the triggers. These experimental results indicate, that the whole FeRh layer cannot ever reach pure AFM phase as an effect of only temperature, which is consistent with previous ab initio calculations [67].

In contrast to the top and bottom sublayers, in the middle (~3rd – 8th) sublayers $X_{\text{AFM}}$ values are dramatically increasing with the decrease of temperature, changing from “fully” FM phase (at 394 K) to a “completely” AFM phase (at 299 K) (Fig. 6. and Fig. 7.).
Fig. 7. $X_{\text{AFM}}$ as a function of temperature and external magnetic field per sublayer, without external electric field.

As the shapes and slopes of the plotted $X_{\text{AFM}}$ - $T$ curves are similar in each middle sublayer (blue lines on Fig. 7.), we can assume that the AFM ↔ FM phase transition occurs (vertically) homogeneously. These findings contradict the results of [41], where the heating induced AFM $\rightarrow$ FM phase transition propagates from top to bottom, but it should be borne in mind, that the substrate, and the thickness of the FeRh layer, was different in that study. In our case, the cause of the homogeneity can be either the homogeneous formation, or the growth of the already existing well distributed FM and AFM domains, during the heating and cooling phases, respectively. As the slope of the $X_{\text{AFM}}$ - $T$ curves is similar and uniform in each sublayer, we can assume, that the phase transition of the alloy is continuous. During mapping of the FeRh surface, similar, continuous, and homogeneous temperature induced AFM / FM phase transition was also found, called Volmer-Webber island type growth [68, 69, 70]. The combination of this previous result (horizontally continuous and homogenous domain growth) with ours (vertically continuous and homogenous domain growth) indicates, that the phase transition of the alloy is homogeneous and continuous in all 3 dimensions throughout the layer (except for the top and the bottom sublayers), if it is triggered by temperature change. From a technology viewpoint, these results showed, that the alloy layer always has to be at least 4 nm thick, in the FeRh/BTO composite, if temperature induced AFM ↔ FM phase transition is required.

3.3.1 Magnetometry control of the temperature induced phase transition of the alloy

The phase transition described in 3.3, is strongly supported by magnetometry. Magnetization was calculated using the substrate area (25 mm$^2$) and the FeRh film thickness (16.8
nm) determined by XRR. The coercive force was extracted, after correcting for a ±0.09 mT sweep-direction dependent offset of the superconducting magnet at every temperature and orientation. It was found, that the (010) magnetization of the $^{57}$FeRh/BaTiO$_3$ composite increases from ~400 emu/cm$^3$ to ~700 emu/cm$^3$ as the temperature increases from 300 K to 400 K (Fig. 8.), which corresponds well to the temperature induced AFM → FM phase transition. The existence of multiple ferromagnetic sublayers (top and bottom sublayers in the GI-NRS model) is also supported by magnetometry, as the hysteresis loop at lower temperatures consists of two separated FM components (Fig. 8.), one with lower, and one with higher coercive force (table 2. in supplementary materials).

Fig. 8. Magnetization, at (010) direction, (A) and coercive force (B) as a function of magnetic field and temperature in FeRh/BTO composite

As the sample is heated up, the coercivity separation decreases (Fig. 8.), which also corresponds well to the GI-NRS determined FM / AFM / FM → FM transition. (Note: the sudden decrease in the coercive force(s) around 400 K can be attributed to the tetragonal → cubic phase transformation of the BTO which forces the coupled FeRh layer into FM phase due to epitaxial strain). At last, the results of magnetometry also shows, that 150 mT external magnetic field, used in the GI-NRS experiments, is enough to reach the full magnetic saturation.

3.4. Depth dependent determination of the phase transition of the alloy as a function of temperature in external magnetic field

The 2$^{nd}$ group of GI-NRS measurements showed, that regardless of the applied external magnetic field, the 1$^{st}$ and 10$^{th}$ sublayers always remain completely in FM phase (Fig. 9.). Therefore, we can assume, that in this composite no temperature can result in a pure AFM phase regardless of whether external magnetic field is applied or not.
Fig. 9. $X_{\text{AFM}}$ as function of depth and temperature, with 150 mT external magnetic field applied, “A” during cooling phase and “B” during heating phase.

The similar shapes and slopes of the $X_{\text{AFM}} - T$ curves in the first two GI-NRS measurement series (Fig. 7.) indicate, that the AFM ↔ FM phase transitions occurred by the same mechanism in both measurement series. Therefore, we can assume, that the application of external magnetic field doesn’t have any localized effect on this composite, but it homogenously preserves the quantity of the FM phase during the cooling phase. However, the comparison of the $X_{\text{AFM}}$ values also reveals, that the AFM phase ratio in the middle sublayers differs depending on the strength of the magnetic field applied during the cooling phase (Fig. 7.), similar to what was previously described for the “average AFM phase ratio” [71]. The difference between the zero-field-cooled and in-field-cooled measurements indicates, that ferromagnetic clusters are always present in the middle FeRh sublayers [72], supporting the previously hypothesized homogeneous domain growth theory instead of the homogeneous domain formation theory (3.3.).

3.5. Depth dependent determination of the phase transition of the alloy as a function of temperature and electric field, in the FeRh/BTO multiferroic

The $X_{\text{AFM}}$ values, that were measured with applied electric field, demonstrated, that in contrast to the effects of temperature, the voltage can even transform the bottom FM sublayer into AFM phase in this multiferroic (Fig. 10.). This different behavior is a consequence of the mechanical strain dependence of the $X_{\text{AFM}}$ phase, which originates from the combination of the
epitaxial coupling between the FeRh and BTO layers and the electric field induced piezoelectric strain of the BTO [73].

Fig. 10. $X_{\text{AFM}}$ as a function of temperature and external electric field per layer, without applied magnetic field

The comparison of the $X_{\text{AFM}} – E$ curves, that were measured at different temperatures (Fig. 10.), revealed, that below 320 K and above 372 K the external electric field has no measurable effect on the AFM / FM phase ratio of the alloy. Below 320 K there is simply not enough FM phase in the alloy for its transformation to be significant. On the other hand, above 372 K the FM phase is too stable to be significantly transformed to AFM phase by the epitaxial coupling. However, within this temperature range, the electric field induced phase transition can easily overpower the effect of temperature in the ~ 9 nm vicinity of the BTO substrate (Fig. 10.). Therefore, this multiferroic coupling is optimal for anticipated devices that can operate with FeRh layer up to 9 nm thick in this temperature range (optimally at 342 K).

In addition, the plotting of $X_{\text{AFM}}$ as a function of depth and $E$ (Fig. 11.) demonstrates, that the electric field induced FM → AFM phase transition occurs practically completely as soon as $E$ reaches a certain limit between 10 kV/m and 30 kV/m. But once the electric field has reached the threshold, its further increase has only a small effect on the phase ratio of the alloy. Overall, phase
transitions with such properties are generally desirable in technical applications (e.g. as magnetic switches).

![Graph](image)

**Fig. 11.** $X_{\text{AFM}}$ as function of depth and electric field, at 331 K temperature, without external magnetic field; during electric field buildup

However, there is also a downside to this electric field effect: contrary to the temperature induced transition, this one is not automatically reversible; i.e. $X_{\text{AFM}}$ does not convert back completely when the electric field drops back to zero. On the other hand, the initial phase ratio of the alloy can be completely restored by heating it and then cooling it back. Notes: 1.: it is even possible, that the electric field induced phase transition also reverses in time, but due to the available beamtime, we could wait only 15 minutes between measurements. 2.: although, we heated the sample straight up to 394 K during this experiment, it is possible that lower temperature is also enough for the reversion.

### 3.6. **Comparison of the depth dependent effects of temperature, external magnetic and electric fields on the phase transition of the alloy**

The results of the 4th GI-NRS experiments showed, that the presence of an external magnetic field reduces the effect of the electric field at all temperatures where it has a measurable effect, although not at the FeRh/BTO interface (Fig. 12.). This inhibition of AFM phase formation is very similar to the effect, that was described during the cooling phase in 3.4. Therefore, in together the four GI-NRS experiments revealed, that the effects of temperature, external magnetic and electric fields on the phase ratio of the alloy in the FeRh/BTO multiferroic may amplify or even eliminate each other (Fig. 12.). However, the relative strength of these effects is not homogenous through the FeRh layer; in the ~4 nm vicinity of the substrate the effect of the electric field is vastly stronger, than the effects of either external magnetic field or temperature (within our experimental conditions). Further away from the interface, the AFM / FM phase ratio is determined by the balance of temperature, magnetic and electric field. In addition, that the effect of the external electric field is sharply terminated by the external magnetic field ~12 nm above the substrate at
352 K; but this cancellation effect extends only up to ~9 nm, if the sample is cooled down to 331 K (Fig. 12.). Therefore, the FeRh thickness, where the external electric field can trigger the FM \(\rightarrow\) AFM phase transition, is determined by the combination of the temperature and the strength of the external magnetic field.

![Image](image_url)

**Fig. 12.** \(X_{AFM}\) as function of temperature, external electric and magnetic fields in the FeRh/BTO composite

4. Conclusion

In summary, the depth dependent effects of temperature, external magnetic and electric fields on the phase ratio of the alloy in the FeRh/BTO multiferroic were determined by GI-NRS measurements.

The FM phase of the top and bottom part of the FeRh layer cannot be ever transformed into AFM phase by temperature alone, regardless of whether magnetic field is present or not. Outside these regions, the decrease of temperature induces the continuous and homogenous growth of the AFM domains in the FeRh alloy, independent on the strength of the external magnetic field. However, the maximum achievable AFM ratio and the temperature of the main phase transition, in turn, depend on an external magnetic field.

In contrast, the applied electric field causes the magnetic phase transition with a strictly different mechanism in the FeRh/BaTiO\(_3\) composite: it triggers a bottom \(\rightarrow\) up oriented mechanical strain, that forces the sudden FM \(\rightarrow\) AFM phase transition after the strength of the electric field reaches a certain limit. By this mechanism the external electric field is capable to transform even the lowest FM sublayer into AFM phase; but at the same time, it also limits the layer thickness, where the voltage can change the phase ratio of the alloy. However, this thickness limit is adjustable by changing the temperature and external magnetic field combination.

From an engineering point of view, the results of this study indicate that for the optimal utilization of the multiferroic coupling of the FeRh/BTO multiferroic, the thickness of the alloy...
should be between 4 and 12 nm; and the devices based on this coupling should be operating around 342 K.

**CRedit authorship contribution statement**

**Attila Lengyel**: Investigation, Data curation, Methodology, Writing - Original Draft. **Gábor Bazsó**: Investigation. **Aleksandr I. Chumakov**: Investigation, Resources. **Dénes L. Nagy**: Investigation, Methodology. **Gergő Hegedűs**: Resources. **Dimitrios Bessas**: Resources. **Zsolt E. Horváth**: Investigation. **Norbert M. Nemes**: Investigation. **Maria A. Gracheva**: Investigation, Writing - Review & Editing. **Edit Szilágyi**: Investigation. **Szilárd Sajti**: Calculation, Software. **Dániel G. Merkel**: Conceptualization, Supervision, Investigation, Data curation, Methodology, Funding acquisition.

**Declaration of Competing Interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

**Data availability**

The data that indicated the above-described results are available in the CONCORDA repository of the Wigner RCP, under entry: (the number will come after this article gets it DOI identifier).

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**Appendix A. Supplementary material**

Supplementary data associated with this article can be found in the online version at (the number will come after it gets its DOI identifier)

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