

INFLUENCE OF MAGNETIC STRUCTURE ON ELECTRIC CHARGE TRANSPORT IN THIN FILMS OF HOLMIUM AND DYSPROSIUM

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High precision electrical resistance measurements were performed on holmium and dysprosium bulk and thin film samples in a wide temperature range from 4.2 K up to the room temperature. Influence of magnetic structure on the resistance *vs* temperature (R *vs* T) curves was observed as a “knee-like” anomaly near the Néel temperature value $T_N = 128.9$ K in Ho bulk sample and below 119.5 K in Ho thin films. Numerical analysis of the R *vs* T data yielded the Curie temperature value $T_C = 19$ K for Ho bulk sample. Unexpected resistance minimum at 9 K and a slope’s change of the R *vs* T curve at ~ 170 K were observed in 215 nm thin Ho film. The X-ray diffraction of Ho films revealed diffraction peaks caused by the hcp structure of Ho and those from a holmium dihydrid. The bulk Dy sample showed a „hump-backed“ resistance anomaly near $T_N = 180.50$ K and a sudden decrease of resistance value at $T_C = 92.00$ K. A “knee-like” resistance anomaly below 179 K was observed in thinner dysprosium films, whereas a “hump-backed” one was observed in thicker films. The X-ray diffraction of Dy films in a thickness range from 25 nm to 350 nm showed peaks from the hcp structure of Dy and small peaks caused by dysprosium dihydrid.

Keywords: holmium, dysprosium, thin films, electrical resistivity, low temperatures, Néel temperature, Curie temperature, X-ray diffraction

1 INTRODUCTION

The discovery of giant magnetoresistance (GMR) just over a decade and a half ago [1, 2], the development of magnetic tunnel junctions with high tunneling magnetoresistance (TMR) [3, 4], discovery of colossal magnetoresistance (CMR) [5] and even newest ballistic magnetoresistance (BMR) [6] is one of the most important experimental facts in magnetic materials in the last decades. GMR was originally observed experimentally in artificially prepared materials-in magnetic Fe/Cr multilayers, later with Co and Ni as ferromagnetic films and with various nonmagnetic spacers like Au, Ag, Cu. As for rare earth metal thin films, magnetoresistance was studied in multilayers with CoSm layers [7], GdCo films [8] and positive GMR was observed in Dy/Sc superlattices [9].

The main technological interest of GMR lies in its ability to detect tiny magnetic fields and, in magnetic recording, for example, to access data much more quickly than conventional inductive read-write heads [10, 11]. Thin film deposition techniques which allow to deposit layers of metals just a few nanometers thick, have transformed GMR into a billion dollar business within a decade of its discovery.

Magnetoelectronic devices, both existing and envisioned, rely on the spin-dependent transport of electrons. The area of the magnetoelectronics, which is at the interface between magnetism and electronics, provides new effects, new capabilities and new functionalities [12, 13]. It promises a wide variety of new devices that combine logic, storage and sensor applications. Moreover, these “spintronic” devices might lead to quantum computers and quantum communication based on solid-state devices, thus changing the perspective of information technology in the 21st century.

Transport and magnetic properties of magnetic metallic thin films therefore exhibit a new considerable renascence since the discovery of spin polarized transport in multilayered ferromagnetic/nonmagnetic structures. It has strongly stimulated both experimentalists and theoreticians to study arrays of magnetic films-see eg [14, 15].

Nevertheless, study of the electron transport properties of thin films of individual materials are of importance for their better understanding. Transition metal thin films like Ni and Cu are also studied [16]. Rare earth metal thin films attracted some time ago the attention of experimentalists and these films are known to exhibit unusual anomalies of their physical properties [17].

The lack of physical properties in the literature stimulated us to study the electrical and structural properties of thin films of holmium and dysprosium. Their parent materials Ho and Dy bulk samples represent the wealth of magnetic structures in the group of lanthanides, which are complicated and appear in the low temperature region [18].

2 EXPERIMENTAL

The investigated thin films were prepared by evaporation onto glass substrates. Holmium films were evaporated in ultrahigh vacuum at $\sim 10^{-7}$ Pa, dysprosium thin films were evaporated in high vacuum of $\sim 10^{-4}$ Pa and covered with a protective SiO layer. High precision electrical resistance measurements of bulk and thin film samples were performed by the conventional four-point dc arrangement using a Keithley stabilized digital programmable current source K 220 and digital nanovoltmeter K 181.

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The temperature of the bulk and thin film samples in the helium cryostat was measured using calibrated Lake Shore Cryoelectronics, Inc. germanium (from 4.2 K to 80 K) and platinum (from 80 K to 300 K) thermometers. Film thickness was measured using the optical Tolansky method.

The crystal structure of Ho and Dy films was determined using the X-ray diffraction with the Bragg-Brentano focusing geometry.

3 RESULTS AND DISCUSSION

3.1 Holmium thin films

Prior to the Ho thin film study, the electrical resistance of the reference Ho bulk sample was measured. The result of this measurement is illustrated in Fig.1 as the R/R_0 vs T dependence (R_0 being the resistance value at 4.2 K and R its value at the temperature T). The residual resistance ratio value (ratio of the resistance at room temperature to that at 4.2 K) of this Ho bulk sample was RRR=18.8.

A “knee-like” resistance anomaly is clearly seen near ≈ 130 K in Fig.1 corresponding to the magnetic phase transition from paramagnetic state to the antiferromagnetic one with spiral structure. The numerical analysis of the resistance data yielded the dR^2/dT^2 vs T dependence with the deep minimum corresponding to the Néel temperature $T_N = 128.94$ K - see the insert in Fig. 1.

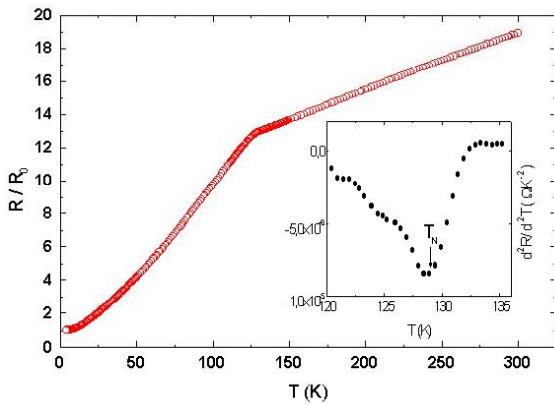


Fig. 1. Temperature dependence of the resistance ratio R/R_0 of Ho bulk sample in the temperature range from 4.2 K to 300 K.. The insert shows the second temperature derivative of resistance near the Néel temperature

The other resistance anomaly near ≈ 20 K, although not clearly seen in Fig. 1, is connected with the magnetic phase transition from the spiral spin structure to the ferromagnetic cone one. But the numerical analysis of the resistance data yielded the dR^2/dT^2 vs T dependence with the minimum at $T_C = 19$ K , which is clearly seen in Fig. 2 and which corresponds to this transition.

The electrical resistance of holmium thin films with thickness of 116 and 215 nm was investigated in the temperature range from 4.2 K up to the room temperature. The result of this investigation is illustrated in Fig. 3 as the resistance ratio vs temperature for these films.

The resistance vs temperature curve of 116 nm thin Ho thin film exhibits a “hump-backed” resistance anomaly near ≈ 120 K, which is connected with the magnetic phase transition from paramagnetic state to the antiferromagnetic one

with spiral structure. The numerical analysis of the experimental data yielded the Néel temperature value $T_N = 119.5$ K.

The qualitatively other result was obtained in 215 nm thin film of holmium. The R vs T dependence shows a “knee-like” anomaly near ≈ 120 K, which resembles to the “knee-like” R vs T anomaly observed for basal plane of holmium single crystal and which is caused by magnetic structure. Moreover, two resistance anomalies were observed in this Ho film - a resistance minimum at 10 K (indicated by T_{min} arrow in Fig. 3 and it is illustrated more clearly in the insert in this Figure 3) and a slope change of the R/R_0 vs T curve indicated by the arrow T_a . The crystal structure study was investigated in Ho films.

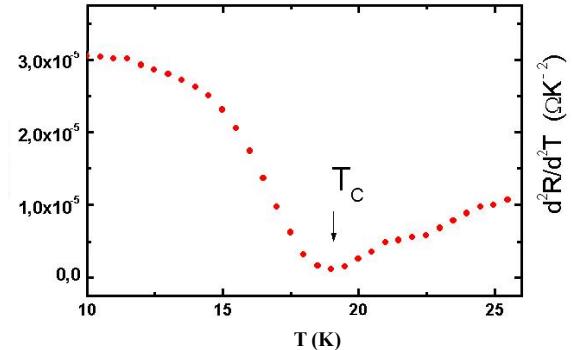


Fig. 2. Second temperature derivative of the resistance of Ho bulk sample below 25 K. The minimum at $T_C = 19$ K corresponds to Curie temperature

It is known that the RE metal thin films readily react with hydrogen to form the dihydrides with the fcc structure [17]. Therefore the positions of expected HoH_2 diffraction peaks were evaluated by means of the Bragg law using X-ray radiation of wavelength $\lambda = 0.17902$ nm and the lattice parameter of the fcc structure of HoH_2 $a = 0.5140$ nm [19]. The diffraction peak caused by (111) plane of the fcc should appear at $2\Theta = 35^\circ$ position (Θ is the Bragg angle).

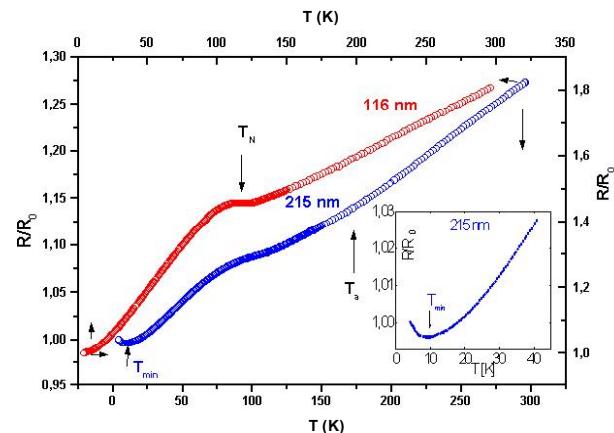


Fig. 3. The resistance ratio R/R_0 vs T curves of two Ho thin films with thickness of 116 nm and 215 nm in the temperature range from 4.2 K to 300 K. The insert shows more clearly the resistance minimum at 9 K . The T_a indicates slope change of the R/R_0 vs T curve at ~ 170 K of the 215 nm Ho thin film..

The diffractogram of 116 nm thin film is shown in Fig. 4. The diffraction peaks at 33.65° , 37.2° and 38.65° are caused by the hcp structure of holmium and the peak at 34.9° is caused by the fcc structure of holmium dihydride.

The position of the observed fcc diffraction peak in this Ho film (34.9°) is in coincidence with the evaluated value (35°), thus we assume, the holmium dihydrid is present in this film. However, comparing the R vs T curves of our Ho bulk sample with those of 116 nm thin film we see that the amount of HoH_2 did not influence qualitatively the observed R vs T dependence of this film.

The diffraction pattern of 215 nm thin film exhibits two peaks at 2Θ angles 36.9° and 38.3° , caused by the hcp structure of holmium, a high peak at 34.9° and a small peak at 40.6° , which are caused by the holmium dihydrid HoH_2 .

We assume, that the larger content of HoH_2 in 215 nm thin film is the reason for the “weaker” resistance anomaly near ≈ 120 K of this film compared with those of 116 nm thin Ho film.

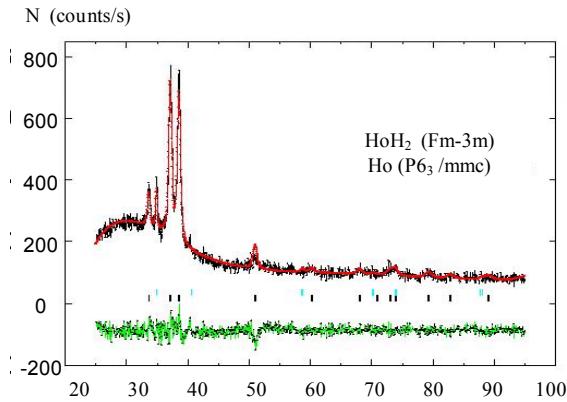


Fig. 4. Retrieved refinement diffractogram for 116 nm Ho film. The upper curve includes experimental points overlaid by the calculated solid line. The lower curve shows the difference between the observed and calculated intensities. Positions of all possible Bragg reflections for the two phases are indicated by vertical marks in the middle

Holmium belongs to rare earths which absorb hydrogen readily and form solid solutions and/or hydrides. The study of the influence of hydrogen on the magnetic and electric properties of monocrystalline thulium showed a variety of the resistance anomalies in TmH_x with H concentration up to $x = 0.1$ in the low temperature region below ~ 60 K, including a resistance minimum, and slope change of R vs T curve in the vicinity of ~ 170 K [20]. The resistance minimum at 9 K and slope change of the R vs T curves at ~ 170 K were also observed in thulium thin films [21]. Thus, both resistance anomalies in 215 nm thin film of holmium could be interpreted, we assume, as caused by hydrogen present in the form of solid solution (α -phase).

3.1 Dysprosium thin films

The electrical resistance of dysprosium bulk sample was investigated as a reference in the temperature range from 4.2 K up to the room temperature. Two anomalies were clearly seen in the R vs T dependence- a “hump-backed” resistance anomaly below 190°K , which is connected with the magnetic phase transition from

paramagnetic state to the antiferromagnetic one with the helical spin structure. and a sudden decrease of the resistance value below ~ 100 K, connected with the transition to the ferromagnetic spin structure [18]. Numerical analysis of the experimental R vs T data yielded the sharp positive minimum of the $d\mu/dT$ vs T curve corresponding to the Néel temperature value $T_N = 180.50$ K and a sharp positive maximum corresponding to the Curie temperature $T_C = 92.00$ K [22].

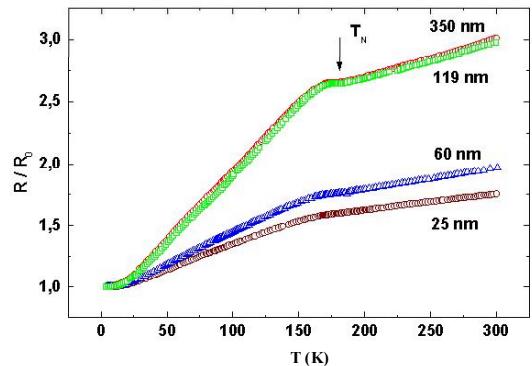


Fig. 5. The resistance ratio R/R_0 vs T dependences of dysprosium films with thickness of 25, 60, 119 and 350 nm in the temperature interval from 4.2 K to 300 K

Unexpected anomalies of the physical properties of dysprosium thin films, mainly the size-effect of resistivity, attracted the attention some time ago and they were reported in the literature-for analysing and explanation see eg [17].

We prepared films of dysprosium in the thickness range from 25 nm to 350 nm. Their electrical resistance was measured in the temperature range from the 4.2 K up to the 300 K. The results of these measurements are illustrated in Fig. 5 for four of them with thicknesses of 25, 60, 119 and 350 nm. The „knee-like“ R vs T anomaly below ~ 180 K for thinner films is represented by 25 and 60 nm thin films and this anomaly is evident in Fig. 5. Thicker films are represented by 119 and 350 nm thin films and these films exhibit a „hump-backed“ resistance anomaly below ~ 180 K, which is evident from Fig. 5. The numerical analysis of the experimental R vs T data of investigated films yielded the Néel temperature values with the result of their decrease from the value $T_N = 179$ K for the 350 nm thin film to the $T_N = 174.4$ K value for the thinnest studied film of 25 nm [17].

The crystal structure of Dy films was estimated by the X-diffraction [17]. A preferential crystal orientation of crystallites in Dy films was observed. The majority of crystallites in thinner films had their (002) planes parallel with the substrate plane. A different preferential orientation of crystallites was observed in thinner films.

Moreover, diffraction lines caused by dysprosium dihydrid DyH_2 were observed in Dy films. Comparing R vs T curves of Dy bulk sample with those of Dy thin films one can see the amount of DyH_2 was in small concentration that did not influence the R vs T curves qualitatively.

4 CONCLUSIONS

We have investigated the influence of the magnetic structure on the electrical resistance of holmium and dysprosium bulk and thin film samples in the temperature interval from 4.2 K up to the room temperature, and the crystal structure of Ho and Dy films by the X-ray diffraction.

From this study following conclusions can be made:

1. Holmium and dysprosium thin films are very sensitive to hydrogen present in vacuum bell-jar and parent material to form dihydrides of these metals and solid solution (α -phase).
2. Observed R vs T dependencies of Ho films (UHV) exhibit anomalies identical with those observed in bulk sample. The T_N values of Ho films are lower than that of the Ho bulk sample. An unexpected additional resistivity minimum at 9 K and the slope change of R vs T curve near \sim 170 K are probably caused by hydrogen present in the form of solid solution.
3. Observed R vs T curves in dysprosium films (HV) exhibit the “knee-like” resistance anomalies near the Néel temperature in thinner films and the “hump-backed” anomalies in thicker films caused by magnetic structure of Dy. The Néel temperature value decreases with decreasing film thickness.
4. The X-ray diffraction studies of Ho thin films revealed their structure with the prevailing Ho hcp phase and the presence of the fcc phase from holmium dihydrid.
5. The X-ray diffraction studies of Dy thin films revealed their structure consists from the prevailing Dy hcp phase and the presence of the fcc phase from dysprosium dihydrid.

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REFERENCES

- [1] BAIBICH, M.N. et al., Phys.Rev.Lett. **61** (1988), 2472
- [2] BINASCH G. et al., Phys.Rev. **B39** (1989), 4828
- [3] MIYAZAKI,T. - TEZUKA, N., J. Magn. Magn. Mater. **139** (1995), L231
- [4] MOODER, J.S. et al., Phys. Rev. Lett. **74**, (1995), 3273
- [5] von HELMHOLTZ R. et al., Phys. Rev. Lett. **71** (1993), 2331
- [6] GARCIA, N. - M.MUÑOZ, M. -ZHAO, Y.W., Phys.Rev.Lett. **82** (1999), 2923
- [7] MIBU, K. - NAGAHAMA, T. - SHINJO, T., J.Magn.Mater.Magn. **156** (1996), 299
- [8] VASKOVSKIJ, V. O. - SVALOV, A. V. -RYAZANTSEV, A. A., J. Magn. Mater. Magn. **156** (1996), 291
- [9] TSUI, F. - UHER, C. - FLYNN, C.P., Phys. Rev. Lett. **72** (1994), 3084
- [10] BARTHÉLÉMY, A. et a., Phys. World, November **1994**, 34
- [11] MATHON, J., Contemporary Physics **32** (1995), 143
- [12] FERT, A. et al., EUROPHYSICS NEWS, **34** (2004), 227
- [13] GRUNDLER, D., Physics World, April **2002**, 39
- [14] GRÚNBERG, P., Acta mater. **48** (2000), 239
- [15] ZHU, H.J. et al. Phys. Rev.Lett. **87** (2001), 016601
- [16] ABDUL,W. - RAZZAQ, M.A., Physica **B253** (19987), 47
- [17] DUDÁŠ, INTRICATE BEHAVIOUR OF RARE EARTH METAL THIN FILMS AT LOW TEMPERATURES, Mercury-Smekal Pu. House, Košice, 2001, 125 p.
- [18] TAYLOR, K.N.R. — DARBY, M.I.: Physics of Rare Earth Solids, Chapman and Hall, London, 1972
- [19] GASGNIER, M. et al., J.Less-Common Met. **34** (1974)
- [20] VAJDA, P. et al., Condensed Matter **1** (1989), 4099
- [21] DUDÁŠ, J. - FEHER, A. - KAVEČANSKÝ, V., J.Alloys and Compounds **278** (1998), 1
- [22] DUDÁŠ, J. - FEHER, A., J.Electr.Engineering **45**, (1994), 84

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