# INFLUENCE OF MAGNETIC FIELD ON ELECTRIC CHARGE TRANSPORT IN HOLMIUM THIN FILMS AT LOW TEMPERATURES

Ján Dudáš <sup>1</sup>, Stanislav Gabáni <sup>2</sup>, Viktor Kavečanský <sup>2</sup>, Andrej Vincze <sup>3</sup>

Department of Theoretical Electrotechnics and Electrical Measurement, Faculty of Electrotechnics and Informatics, Technical University, Park Komenského 3, 043 89 Košice Phone: 055/602 2706, fax: 055/63 301 15, E-mail: Jan.Dudas@tuke.sk
<sup>2</sup> Institute of Experimental Physics, Slovak Academy of Sciences, 040 01 Košice, Slovakia
<sup>3</sup> International Laser Center, Ilkovičova 3, 812 19 Bratislava

**Summary** Holmium thin films were prepared by evaporation in ultrahigh vacuum (UHV) and high precision electrical resistance measurements were performed on them as well as on holmium bulk sample in the wide temperature range from 4.2 K up to the room temperature. Electric charge transport is profoundly influenced by the magnetic structure at low temperatures and a "knee-like" resistance anomaly was observed near the transition from paramagnetic state to basal-plane spiral structure in bulk with the Néel temperature  $T_N = 128.9 \text{ K}$  and below ~ 122 K in thin Ho films in a thickness range from 98 nm to 215 nm. Unexpected resistance minimum at ~ 9 K and a slope's change of the R vs. T curve near ~ 170 K was observed in 215 nm thin film. Application of magnetic field parallel to the substrate and thin film plane for temperatures below ~ 150 K caused the decrease of resistance value with increasing magnetic flux density. Increasing suppression of the  $T_N$  value up to ~ 5 K with increasing flux density value up to 5 T was observed in Ho films. The X-ray diffraction of Ho films revealed diffraction peaks originating from the h.c.p. structure of Ho and those from holmium dihydride. The secondary ion mass spectroscopy (SIMS) showed the presence of hydrogen, holmium dihydrid and  $Ho_2O_3$   $Ho_3O_5$ . Organic components related to  $C_xH_y$  ions, species of Fe, Cu, Co, Ca, Hf and contaminants Na, K were identified in holmium films.

### 1. INTRODUCTION

The discovery of giant magnetoresistance (GMR) just over a decade and a half ago, the development of magnetic tunnel junctions with high tunneling magnetoresistance (TMR) [1], discovery of colossal magnetoresistance (CMR) [2] and even newest ballistic magnetoresistance (BMR) [3] 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 [4], GdCo films [5] and positive GMR was observed in Dy/Sc superlattices [6].

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 [7]. 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 funcionalities [8, 9]. 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 21 st 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 e.g. [10, 11].

Neverheless, 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 [12]. 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 [13].

The lack of physical properties in the literature stimulated us to study the electrical and structural properties of thin films of holmium. Their parent material Ho bulk sample represents the wealth of magnetic structures in the group of lanthanides, which are complicated and appear in the low temperature region [14].

### 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. 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.

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 films was determined using the X-ray diffraction with the Bragg-Brentano focusing geometry.

The purity and composition of the samples was studied using secondary ion mass spectroscopy. We employed a time of flight based SIMS instrument (Ion-TOF, SIMS, IV) with high energy Au<sup>+</sup> primary source. For the depth profiling of the structure the high energy pulsed primary gun combined with a low energy sputter gun  $(O_2^+)$  because of low erosion rate. The sputtering ion beam was rastered over the area of  $300x300~\mu m$  while the primary beam is rastered within the  $100x100~\mu m$  area in the center of sputtered area. The high resolution TOF detection system was performed by a single counting using a microchannel plate detector, which has parallel mass detection in the range up to  $10.000~\mu m$  [15].

### 3. RESULTS AND DISCUSSION

Prior to the Ho thin film study, the electrical resistance of the reference Ho bulk sample was measured. 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 was observed near  $\approx$  ~ 130 K in 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  $T_{\rm N}=128.94$  K. The transition to other magnetic phase - to ferromagnetic spin structure was obtained by the

second temperature derivative of the resistance with the result  $T_C = 19 \text{ K}$ .

The electrical resistance of holmium thin films with thickness of 98 nm to 215 nm was investigated in the temperature range from 4.2 K up to the room temperature and in zero magnetic field. The result of this investigation is illustrated in Fig. 1 for two of them as the resistance ratio vs. temperature.

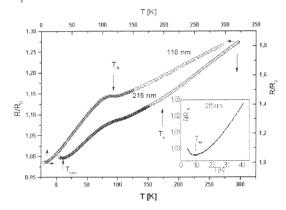


Fig. 1. The resistance ratio  $R/R_0$  vs. T dependences of two Ho films with thickness of 116 nm and 215 nm in the temperature range from 4.2 K up to 300 K. The  $T_a$  indicates slope's change at  $\sim$  170 K of the 215 nm Ho film. The insert shows more clearly the resistance minimum at 9 K in this film.

The resistance vs. temperature dependence of 116 nm thin Ho thin film exhibits a "hump-backed" resistance anomaly near  $\approx$  120 KThe numerical analysis of the experimental data yielded the Néel temperature value  $T_{\rm N}$  = 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  $\approx 120$  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  $T_{\rm min}$  arrow in Fig. 1 and it is illustrated more clearly in the insert in this figure) and a slope's change of the R /  $R_0$  vs. T dependence near  $\sim 170$  K, indicated by the arrow  $T_a$ .

The electrical resistane of Ho films was investigated in the magnetic field up to 5 T in the temperatures below  $\sim 150$  K. The result of this investigation is illustrated in Fig. 2 for 196 mnm thin Ho film. The influence of magnetic field on electric charge transport is evident in this figure as a decrease of the resistance value with increasing magnetic field. Moreover, incresing suppression of the  $T_N$  value up to  $\sim 5$  K with increasing magnetic field up to 5 T was observed.

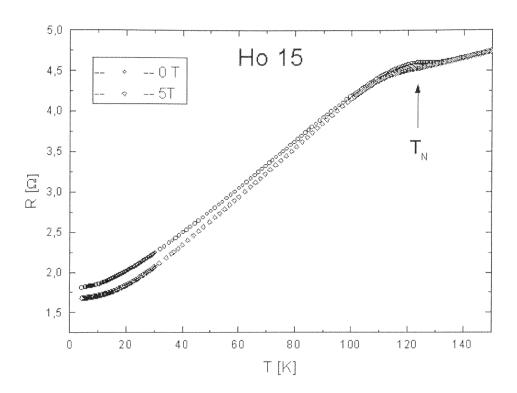


Fig.2. Temperature dependence of the resistance R of Ho thin film sample with thickness of 196 nm in zero magnetic field and in magnetic field with 5 T in the temperature range from 4.2 K to 150 K. The decrease of the resistance value due to magnetic field is evident in this figure.

The crystal structure study was investigated in Ho films. It is known that the RE metal thin films readily react with hydrogen to form the dihydrides with the fcc structure [13, 16]. Therefore the positions of expected HoH<sub>2</sub> diffraction peaks were evaluated by means of the Bragg law using X-ray radiation of wavelength  $\lambda = 0.17902$  nm and the lattice parameter a = 0. 5140 nm [16] of the fcc structure of HoH<sub>2</sub>. The diffraction peak caused by (111) plane of the fcc phase should appear at 2  $\Theta$  = 35  $^0$  position ( $\Theta$  is the Bragg angle).

The diffractogram of 116 nm thin film is shown in Fig.3. The diffraction peaks at  $33.65^{-0}$ ,  $37.2^{-0}$  and  $38.65^{-0}$  are caused by the hcp structure of holmium and the peak at  $34.9^{-0}$  is caused by the fcc structure of holmium dihydride.

The position of the observed fcc diffraction peak in this Ho film  $(34.9^{\circ})$  is in coincidence with the evaluated value  $(35^{\circ})$ , 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<sub>2</sub> did not influence qualitatively the observed *R* vs. *T* dependence of this film.

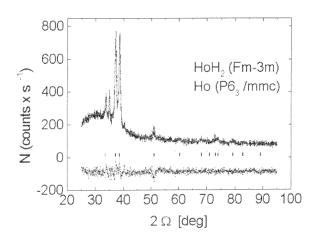


Fig.3. The Rietveld refinement diffractogram for 116 nm Ho film. The upper curve includes experimentall points overlayed by the calculated solid line. The lower curve shows the difference between the observed and calculated intensities. Positions of all possible Bragg reflections for the two phases are indicated by vertical marks in the middle.

The diffraction pattern of 215 nm thin film exhibits two peaks at  $2\Theta$  angles  $36.9^{-0}$  and  $38.3^{-0}$ , caused by the hcp structure of holmium, a high peak at  $34.9^{-0}$  and a small peak at  $40.6^{-0}$ , 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  $\approx 120~K$  of this film compared with those of 116 nm thin Ho film.

Holmium belongs to rare earths which absorb hydrogen readily and form solid solutions and/or hydrides. The the influence of hydrogen on the magnetic and electric properties was studied in other lanthanide-monocrystalline thulium. This study showed a variety of the resistance anomalies in  $TmH_x$  (H concentration up to x = 0.1) in the low temperature region below ~ 60 K, including a resistance minimum and slope's change of the R vs. T curve in the vicinity of ~ 170 K [17]. The resistance minimum at 9 K and slope's change of the R vs. T curves at ~ 170 K were also observed in thulium thin films [18]. 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 ( $\alpha$ -phase).

The SIMS spectra were taken from the 196 nm Ho film, as well as from the parent Ho bulk sample and from the glass carrier of the Ho film. The positive secondary ion mass spectra of the Ho film showed the film is homogeneous with a plane surface. The H and Ho signal in the Ho film on glass were constant until the interface. The signals from  ${\rm HoH_2}$ ,  ${\rm Ho_2O_3}$ ,  ${\rm Ho_3O_5}$  were present together with signals from other impurities—organic components related to  ${\rm C_xH_y}$  ions, and some other species like Fe, Cu, Ca, Co, Hf originating from parent Ho bulk, and Na, K from the glass substrate, are identified.

#### 4. CONCLUSIONS

We have investigated the influence of magnetic field on the electrical resistance of holmium thin film samples in the wide temperature interval, the crystal structure of Ho films by the X-ray diffraction and the purity of Ho films using SIMS .

From this study following conclusions could be made:

- 1. Holmium thin films are very sensitive to hydrogen present in vacuum bell-jar and parent material to form dihydrides, and solid solution of H ( $\alpha$ -phase).
- 2. Observed R vs. T dependencies of Ho films (UHV) exhibit anomalies identical with those observed in bulk sample. The T<sub>N</sub> values of Ho films are lower than that of the Ho bulk sample. An unexpected additional resistivity minimum at 9 K and the slope's change of R vs. T curve near ~ 170 K are probably caused by hydrogen present in the form of solid solution.

- 3. 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.
- 4. The SIMS depth profile spectra detected signals from Ho, holmium dihydrid,  $\text{Ho}_2\text{O}_3$   $\text{Ho}_3\text{O}_5$ . Organic components related to  $\text{C}_x\text{H}_y$  ions, other species like Fe, Cu, Ca, Co, Hf and Na, K contaminants are identified in the Ho layer.

## Acknowledgement

The research was supported by the Slovak Scientific Grant Agency VEGA under Projects 1/0163/0, 2/4061/04 and 1/0130/03 and by the contract I/2/2003 of the Slovak Academy of Sciences for the Centres of Excellence.

#### REFERENCES

- [1] Mooder, J.S. et al., Phys. Rev. Lett. 74, (1995), 3273
- [2] von Helmholt R.et al., Phys. Rev. Lett. **71** (1993), 2331
- [3] Garcia, N. M.Muñoz, M. -Zhao, Y.W., Phys.Rev.Lett. 82 (1999), 2923
- [4] Mibu, K. Nagahama, T. Shinjo, T., J.Magn.Mater.Magn. 156 (1996), 299
- [5] Vaskovskij, V. O. Svalov, A. V. -Ryazanttsev, A. A., J. Magn. Mater. Magn. 156 (1996), 291
- [6] Tsui, F. Uher, C. Flynn, C.P., Phys. Rev. Lett. **72** (1994), 3084
- [7] Mathon, J., Contemporary Physics 32 (1995), 143
- [8] Fert, A. et al., EUROPHYS. NEWS, 34 (2004), 227
- [9] Grundler, D., Physics World, April 2002, 39
- [10] Grünberg, P., Acta mater. 48 (2000), 239
- [11] Zhu, H.J. et al. Phys. Rev. Lett. 87 (2001), 016601
- [12] Abdul, W. Razzaq, M.A., Physica B253 (1998), 47
- [13] Dudáš, J. INTRICATE BEHAVIOUR OF RARE EARTH METAL THIN FILMS AT LOW TEMPERATURES, Mercury-Smékal Pu. House, Košice, 2001, 125 p.
- [14] Taylor, K.N.R. Darby, M.I.: Physics of Rare Earth Solids, Chapman and Hall, London, 1972
- [15] J. C. Vickerman, D. Briggs: TOF-SIMS: Surface analysis by mass spectrometry, IM Publications and Surface Spectra Ltd, UK, 2001
- [16] Gasgnier, M. et al., J.Less-Comm. Met. 34 (1974),
- [17] Vajda, P. et al., Condensed Matter 1 (1989), 4099
- [18] Dudáš, J. Feher, A. Kavečanský, V., J. Alloys and Compounds 278 (1998), 1