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ON INFLUENCE OF MAGNETIC STRUCTURE ON THE ELECTRIC CHARGE TRANSPORT IN SAMARIUM AND THULIUM THIN FILMS[1]

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Abstract

The temperature dependence of the electrical resistivity of samarium and thulium thin films has been studied in the temperature range from 4.2 K to 300 K. The influence of magnetic structure on the resistivity is clearly seen on the obtained resistivity vs. temperature dependences of Sm films prepared by evaporation in high vacuum as well as that of Tm films prepared in UHV. The obtained values of Néel temperatures are lower than those of bulk samples and decrease with decreasing film thickness. Residual resistance ratio exhibits similar behaviour as Néel temperature.

Keywords: rare earth metals, rare earth thin films, electrical conductivity, magnetic phase transition.

Introduction

The group of rare earth metals (REM) exhibits probably more interesting features than any other series of elements. These include a somewhat exotic structural behaviour and wealth of magnetic structures. The behaviour of the electrical resistivity and other transport properties is dominated by the contributions which have their origin in the various spin configurations of ferro- and antiferromagnetic states that occur mainly at low temperatures.

Thin films containing REM are studied as promising for technical applications, e.g. REM-TM (transition metals) films as materials for data storage or HTS films for electronics — as Josephson devices and chip interconnectors, the GdBaCuO films on Si substrates without buffer layer.

Thin films of REM have awoken considerable interest in the last decade and a half [1, 2]. The size-effect of the electrical resistivity at room temperature has been studied in these films particularly. Electrical and magnetic properties at low temperatures have been studied mostly in Dy films.

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The aim of this paper is to refer to the influence of the magnetic structure on electric charge transport in Sm and Tm thin films at low temperatures.

Magnetic Structures of Sm and Tm

We concentrated on Sm and Tm, the magnetic structures of which represent their wealth in REM group. The crystal structure of Sm is quite complicated consisting of nine-layer stacking sequence ABABCBCACA of three A, B, C hexagonal close packed layers [3]. The B and C sites have hexagonal and A sites cubic near neighbour environments. Neutron diffraction studies revealed antiferromagnetic arrangement of spins in hexagonal sites below the Néel temperature $T_N^h = 106\,K$, and spins in cubic positions are arranged antiferromagnetically below the Néel temperature $T_N^c = 13.8\,K$.

Thulium exhibits h.c.p. crystal structure. The magnetic moments order in c–axis modulated (CAM) magnetic structure below $T_N = 56\,K$ which gradually squares up at lower temperatures. At 32 K a ferromagnetic component appears and at lowest temperatures an anti-phase 'square-wave' structure is obtained with four layers having the spins 'up' and the next three layers with spins 'down'.

The situation in the vicinity of magnetic phase transition is complicated by the coexistence of magnetic structures and also by some temperature hysteresis associated with these transitions.

Experiment Description

Samarium bulk sample was cut from a rod with a diameter $\phi = 5\,mm$ and a length of 5 cm and thinned to $\phi = 2.1\,mm$ with 19 mm distance of potential probes. Thulium bulk sample had a form of paralleloid with the dimensions of $21.65 \times 1.27 \times 0.28\,mm^3$. Thin silver current leads and potential probes were attached by electric discharge method.

Thin films of Sm of various thicknesses were prepared by evaporation from the other part of bulk samarium rod onto pre-cleaned glass substrates in the high vacuum coating unit. Liquid nitrogen trap was used in order to avoid the contamination of thin films with water vapour and we did not allow condensation on the substrate until the gettering action was finished. Thus, the resulting pressure in the bell-jar was $\sim 1 \times 10^{-4}$ Pa. Prior to the deposition the silver electric contacts were deposited. Silver current leads and potential probes were cemented at appropriate positions using silver paint. To avoid the contamination with air, thin films were covered with SiO layer.

As for thulium films, we have prepared them in the like manner as Sm films [4]. However, the anomalies of their resistivity vs. temperature dependences were different from those of pure bulk Tm and the influence of the magnetic structure was overlapped by another conduction electron scattering mechanism. The interpretation of the influence of hydrogen in thulium crystal lattice was experimentally proved later [5]. Therefore, afterwards Tm thin films were prepared by evaporation in the ultrahigh vacuum $\sim 10^{-7}$ Pa onto pre-cleaned glass substrates heated up to $\sim 250°C$, the other steps were made as by Sm films.

Film thickness was measured by optical Tolansky method.

A conventional four probe d.c. arrangement was used to measure the temperature dependence of electrical resistance of bulk in the helium cryostat [6] using digital programmable current source Keithley 220 and digital nanovoltmeter Keithley 181. To avoid the influence of thermal e.m.f. and contact potentials, the current was commutated and the average value of voltage drop across the sample was used for resistance evaluation. The accuracy of resistance measurement was 0.01%.

The temperature of the films in the cryostat was measured by calibrated Lake Shore Cryotronics, Inc. germanium (from 4.2 K to 80 K) and platinum (from 80 K to 300 K) thermometers.

Results and Discussion

Prior to the thin film study we measured resistance (R) vs. temperature dependences of Sm and Tm bulk samples that have been used as reference samples.

Samarium

Samarium bulk sample 99.9% pure (from GIREDMET, Moscow), of which thin Sm films were prepared, was studied in the temperature interval from 4.2 K to 300 K. Two anomalies of R vs. T dependence are evident: 'knee-like' anomaly below ~ 110 K and a rapid decrease of R value with decreasing temperature below ~ 14 K. The values of magnetic transitions we obtained using numerical analysis described in [7] with the result $T_N^h = 106.8$ K and $T_N^c = 13.4$ K. Residual resistance ratio (RRR) of this sample was 12.

Thin Sm films were investigated in the thickness range from 37 nm to 115 nm. Typical resistivity (ρ) vs. temperature (T) dependences of two of them are illustrated in *Fig. 1* with clear depiction of both types of anomalies. Numerical analysis of the experimental data by the above method yielded:

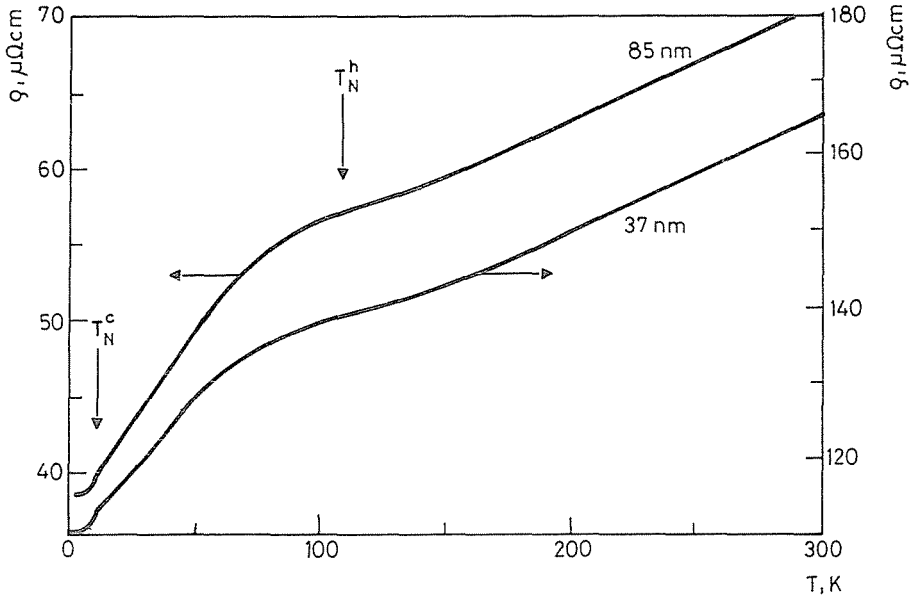

Fig. 1. The typical electrical resistivitys. temperature dependences of two samarium thin films

T_N^h values of all studied films are lower than those of bulk and T_N^h value decreases with decreasing film thickness, being 105.2 K for 115 nm thin film and 102.5 K for 37 nm thin film. T_N^c values of all the films are lower than those of bulk and slightly decrease with decreasing film thickness being 11.9 K for 115 nm thin film and 9.0 K for 37 nm thin film.

RRR value of all films was lower than that of bulk, and decreases with decreasing film thickness from $RRR = 2.12$ for 115 nm thin film to $RRR = 1.54$ for 37 nm thin film.

Thulium

Thulium bulk sample from GIREDMET, Moscow was treated by vacuum distillation method in order to obtain higher purity with the result of $RRR = 145$. This sample was also studied in the mentioned temperature range. The 'hump-backed' anomaly of R vs. T dependence connected with the transition from paramagnetic to antiferromagnetic state was observed below $\sim$ 60 K. The local extreme of the $d\rho/dT$ vs. T dependence by the

method [7] yielded the Néel temperature value $T_N = 57.5$ K. Thin thulium films were investigated in the thickness range from 16 nm to 370 nm. Typical temperature dependences of the ratio of resistivities at temperature T and at 4.2. K are illustrated in *Fig. 2* for four films — 16 nm (1), 89 nm (2), 189 nm (3) and 364 nm (4). 'Hump-backed' anomaly for 364 nm thick film and a change of the slope for 16 nm, 89 nm and 189 nm thin films, corresponding to the paramagnetic–antiferromagnetic transition, are clearly seen in this figure. Temperature dependences of resistivity for thinner films resemble those found for the basal plane of Tm single crystal, whereas 'hump-backed' anomaly found in thicker films is similar to that found for the single crystal c-axis [3]. This suggests the preferential orientation of crystallites in our Tm films.

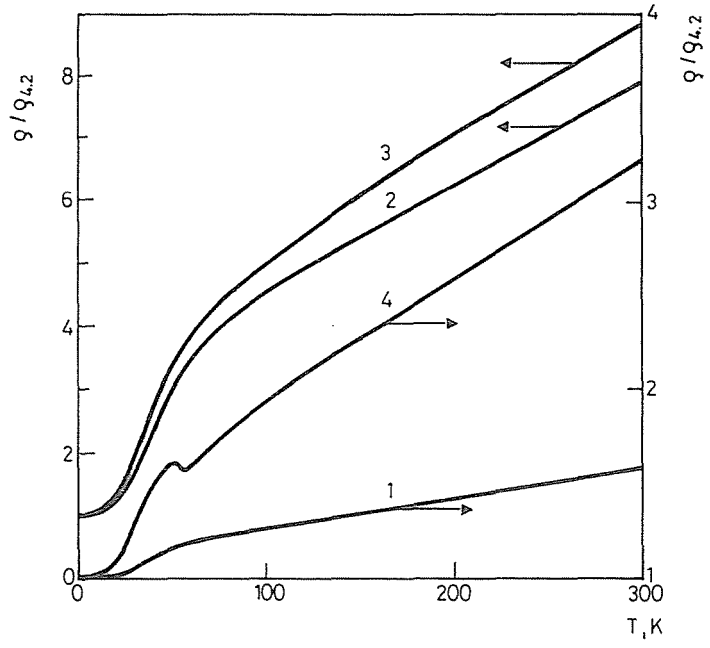

Fig. 2. The typical temperature dependences of the electrical resistivity of four thulium thin films

Numerical analysis yielded Néel temperature values, which are lower than that of bulk for all the studied films, and T_N value drops with decreasing film thickness being 54.0 K for 364 nm thick film and 49.0 K for 16 nm thin film.

RRR value of all Tm films is lower than that of bulk, and drops with decreasing film thickness from $RRR = 3.2$ for 364 nm thin film to $RRR = 1.57$ for 16 nm thin film.

Influence of Thickness on Magnetic Phase Transition

We assume that one of the following reasons or their combination have caused the observed T_N vs. t dependence:

1. The decrease of T_N^h value was caused by increasing internal stresses with decreasing film thickness. The presence of interval stresses was experimentally proved for metal thin films prepared on non-metal substrates (see e. g. [8]), and the decrease of magnetic phase transition temperature values with increasing pressure acting on rare earth metals was experimentally proved (see in [3]). Our thin films were evaporated onto glass substrates and SiO layer, evaporation onto the other film surface could have caused also the stresses in thin film.
2. The decrease of magnetic phase transition temperature value could be caused by the increasing relative contamination of films with decreasing film thickness. Such behaviour was experimentally observed, e. g. for bulk Dy [9]. RRR value is influenced also by contamination and decreases with decreasing thickness of our films.
3. The decrease of T_N value was observed with decreasing film thickness as predicted by theories (e. g. [10]).

We can assume from the comparison of RRR vs. t and T_N vs. t dependences of our films that the second mechanism could be prevailing.

The observed decrease of the Néel temperature in Sm and Tm films is in accordance with other magnetic transition metals and also with Dy films [11].

Conclusions

We have studied the influence of magnetic structure on the electric charge transport in samarium and thulium thin films in the wide temperature range from 4.2 K to 300 K. The following conclusions could be drawn from this study:

1. The magnetic structure influences the electric charge transport in these films in the studied range of thicknesses profoundly, and in the same manner as by bulk samples.
2. The magnetic phase transition temperature values of Sm and Tm films are lower than that of bulk sample and their value decreases with decreasing film thickness.

3. Residual resistance ratio values of our films are lower than those of bulk samples and RRR value decreases with decreasing film thickness.

Further study, mainly study of influence of stresses on magnetic phase transition temperature value in very pure rare earth metal thin films is needed for full clarification of this topic.

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