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Magnetic properties and magnetocaloric effect in ferrimagnetic Gd/Co multilayers

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This work presents the results of a study of magnetic and magnetocaloric properties of Gd/Co multilayer ferrimagnetic films in the temperature range including the magnetic compensation temperature T_{comp}. A strong magnetic field shifts the value of T_{comp} to the region of higher temperatures, which is associated with the appearance of a non-collinear magnetic structure near T_{comp}. It was found that the sign of the magnetocaloric effect changes when passing through the compensation temperature. An increase in the amplitude of the external magnetic field suppresses the change of the sign of the magnetocaloric effect.

Keywords: magnetic multilayered films, ferrimagnets, magnetocaloric effect, non-collinear magnetic structures.

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1. Introduction

Investigation of magnetocaloric materials in thin-film state started approximately 30 years ago [1]. Interest in these investigations is caused both by the ability to track the influence of the size factor on the magnetocaloric effect (MCE) and by the expected development of low-dimensional magnetic cooling devices [2,3]. The largest body of literature is dedicated to heavy rare earth element (Gd, Tb, Dy, Ho) films [4–12] and ferrimagnetic alloys thereof with Co [13–16]. Moreover, Gd/SFM type multilayer structures, where SFM are ferromagnetic layers with high Curie temperature, were used to show that, due to the interlayer exchange interaction, magnetic cooling efficiency in such systems may be much higher than that for homogeneous magnets [17–20]. It is also known that MCE sign reversal is observed in ferrimagnetic materials near the magnetic compensation state [21,22]. This work describes the results of study of magnetic an magnetocaloric properties of ferrimagnetic Gd/Co multilayer films in the temperature region including the magnetic compensation temperature.

2. Research technique

$[\text{Gd/Co}]_6$ multilayer films were deposited on glass substrates by magnetron sputtering of the appropriate targets in argon. Thickness of all layers was 7 nm. To prevent oxidation, Gd/Co multilayer films were protected by 5 nm buffer and top W layers. The films were deposited in the 0.02 T permanent magnetic field oriented in the substrate plane. Sample microstructure data was obtained by the X-ray diffraction method (PHILIPS X'PERT PRO diffractometer, CuK_α). To evaluate the quality and real thickness of the layers, X-ray reflectometry (XRR) was performed using the Rigaku Smartlab diffractometer. Magnetic measurements were made using the PPMS DynaCool 9T measuring system.

3. Findings and discussion

The XRD patterns show two clear peaks (Figure 1, *a*). One of them was identified as corresponding to the (002) line of the HCP lattice of Co (hcp (002)), and the second peak may be a superposition of two peaks, one of which corresponds to the (100) line of the HCP lattice of Co (hcp (100)), and the second one corresponds to the (110) line of the BCC lattice of W (bbc (110)). The mean crystallite size was calculated using the Scherrer equation by the Co peak (hcp (002) parameters and was equal to 5 nm for the Co layers. The absence of other lines in the XRD pattern indicates that the Gd layers are in the X-ray amorphous state, i.e. the mean size of Gd crystallites is 2 nm max. The experimental XRR pattern of the $\text{W(5 nm)/[Gd(7 nm)/Co(7 nm)]}_6\text{/W(5 nm)}$ film was adequately described using nominal layer thicknesses (Figure 1, *b*). RMS roughness of the Gd/Co interlayer boundaries was 1 nm.

Field dependences of magnetization $M(H)$ measured in different directions in the sample plane showed that induced magnetic anisotropy was formed in the films where the easy axis (EA) orientation coincides with the magnetic field orientation that existed in the substrate plane during the film structure sputtering process. All further measurements described in the work were performed with magnetic field oriented along EA.

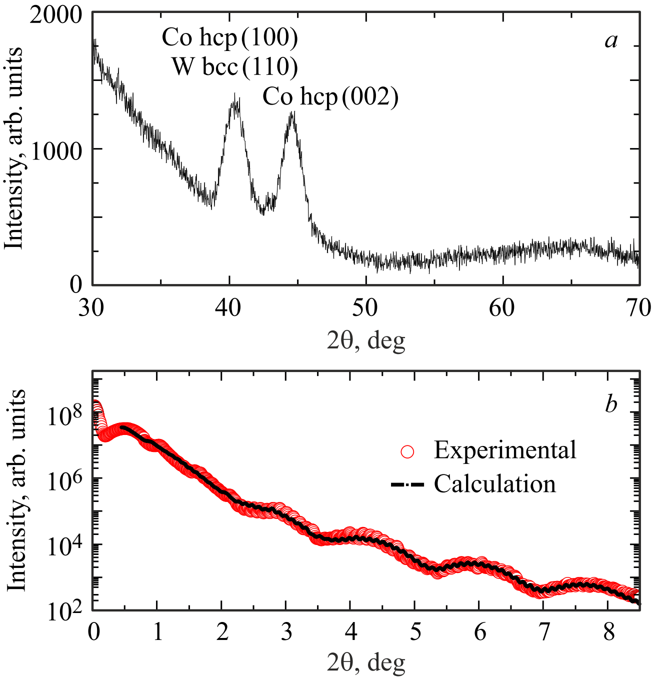

Figure 1. XRD pattern (a) and XRR pattern (symbols) and calculation data (solid line) (b) for the W(5 nm)/[Gd(7 nm)/Co(7 nm)]$_6$/W(5 nm) film.

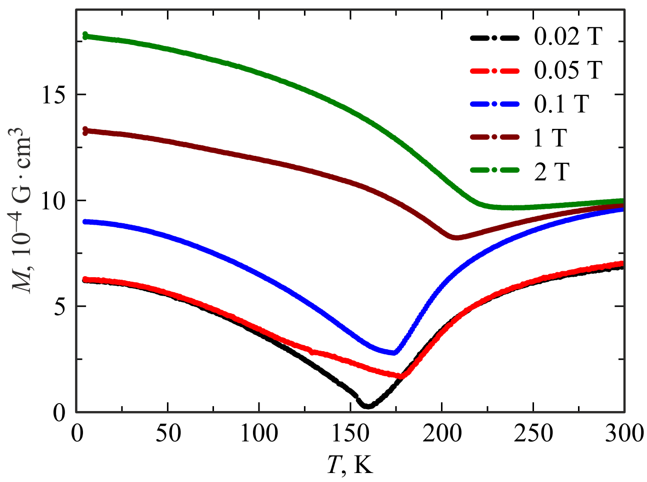

Figure 2. Magnetization-temperature dependence of the [Gd/Co]$_6$ film measured at various external magnetic field strengths.

$M(T)$ of the [Gd/Co]$_6$ film has a ferrimagnetic-specific minimum near the compensation temperature $T_{\mathrm{comp}} \approx 165$ K (Figure 2).

At $\mu_0 H = 0.02$ T, the magnetization almost vanishes at $T = T_{\mathrm{comp}}$. This may be treated as indirect evidence of chemical and structural homogeneity of the [Gd/Co]$_6$ layers [23]. $M(T)$ measured at other field strengths show that an increase in the measurement field leads to smearing of the minimum, and the magnetization doesn't reach zero, which indicates that antiparallel ordering of the magnetic moments of the Gd and Co layers is lost and a noncollinear magnetic structure appeared in a strong magnetic field [24,25]. Typical kinks of the magnetization curves are an evidence of such magnetic structure appearing when the field increases above some critical H_{cr} (Figure 3). It can be seen that at the approach to T_{comp}, H_{cr} decreases as expected.

Variation of the magnetic part of entropy ΔS_M was determined using the magnetic isotherm measurements based on the Maxwell ratio:

$$\Delta S_M = -\int_{H_2}^{H_1} \left(\frac{\partial M}{\partial T}\right) dH, \qquad (1)$$

where H is the magnetic field, M is the magnetization, T is the temperature. Magnetic isotherms were measured at 5 K temperature intervals with magnetic field variation up to 7 T. Temperature dependences $-\Delta S_M(T)$ for different field variation amplitudes are shown in Figure 4.

At $\mu_0 H < 5$ T, when T_{comp} is crossed, the sign of $\Delta S_M(T)$ is reversed on $-\Delta S_M(T)$. As mentioned above, the [Gd/Co]$_6$ films are artificial ferrimagnetic materials and the behavior of $-\Delta S_M(T)$ may be described similar to the bulk ferrimagnetic material [26]. At $T < T_{\mathrm{comp}}$, the magnetic moment of the Gd layer subsystem is co-directional with the external magnetic field, therefore a „ferromagnetic“ type paraprocess takes place in the Gd layers and the field reduces the magnetic part of entropy, while at $T > T_{\mathrm{comp}}$, where the magnetic moments of the Gd layers and the external magnetic field are antiparallel, an „antiferromagnetic“ type paraprocess appears, the external field destroys the magnetic order in the Gd layers and increases the magnetic part of entropy. A similar situation takes place with the magnetic subsystem of the Co layers, but in the reverse order with respect to T_{comp}. However, in the given temperature range, the Gd layer subsystem has a more pronounced magnetization-temperature dependence

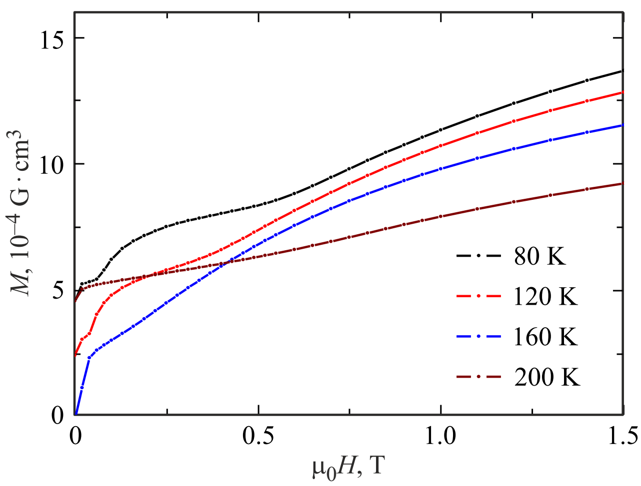

Figure 3. Magnetization curves of the [Gd/Co]$_6$ film measured at different temperatures.

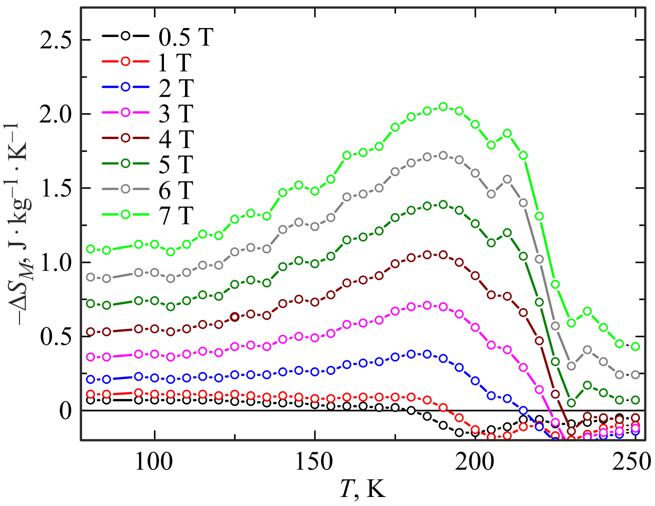

Figure 4. Temperature dependences of the variation of the magnetic part of entropy of the $[Gd/Co]_6$ film for different external magnetic field amplitudes.

and is in a more disordered state than the Co layers, therefore, the external field acting on the subsystem induces a large paraprocess , and, consequently, a large MCE higher than MCE associated with the magnetic system of the Co layers. Therefore, sign reversal on $-\Delta S_M(T)$ near T_{comp} is caused by the MCE sign reversal of the Gd layer subsystem.

In bulk ferrimagnetic materials and RE–TM (rare earth–transition metal) films, MCE sign is reversed almost at $T = T_{comp}$ [15,26]. In our case $T_{comp} \approx 165$ K, if T_{comp} is defined as the temperature of the minimum on $M(T)$ at $\mu_0 H = 0.02$ T (Figure 2). However, the MCE sign is reversed at higher temperatures, and the sign reversal temperature increases with H (see Figure 4). This is due to the fact that occurrence of a noncollinear magnetic structure near T_{comp} in a strong magnetic field shifts T_{comp} to the higher temperature region (Figure 2). At $\mu_0 H \approx 5$ T and higher, evolution of the noncollinear phase, when T_{comp} is crossed, probably takes place in such a way that vast majority of the field component of the Gd layer magnetization is co-directional with the external field. This leads to almost complete disappearance of the minimum on $M(T)$, and the shape of $-\Delta S_M(T)$ becomes similar to the dependence for ferromagnetic materials near the Curie temperature (Figures 2 and 4).

4. Conclusion

In the ferrimagnetic Gd/Co films, magnetocaloric effect sign reversal is observed when the compensation temperature is crossed. MCE behavior in the T_{comp} region is defined mainly by the magnetic moment of the Gd layer subsystem similar to crystalline ferrimagnetic materials and amorphous ferrimagnetic RE–TM films containing the magnetic Gd subsystem. Occurrence of the noncollinear magnetic phase near T_{comp} in the external magnetic field is the main cause defining the behavior of the temperature dependence of the variation of the magnetic part of entropy for the Gd/Co multilayer films.

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Conflict of interest

The authors declare that they have no conflict of interest.

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