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Engineered Gd-Co based multilayer stack to enhanced magneto-caloric effect and relative cooling power

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Magnetic refrigeration based on the magneto-caloric effect is one of the best alternatives to compete with vapor-compression technology. The viability of a magnetic refrigeration system for magnetic cooling can be tested by exploiting the materials in various forms, ranging from bulk to nanostructured materials. In order to achieve a wide refrigerating temperature range in magnetic refrigeration, we study in this paper a 100 nm-thick Gd-Co alloys-based multilayer stack. The stack is made of four individual Gd-Co alloy layers with different values of concentration and Curie temperature (T_C). A magnetic entropy change associated with the second-order magnetic phase transition was determined from the magnetic isotherms. Moreover, the relative cooling power (RCP) of the studied Gd-Co-based multilayer is enhanced compared to the one of bulk Gd, and reaches a value of 200 J/kg. Such an enhancement of the RCP is not due to an enhanced maximum variation of entropy, but this is due to a much broader magnetic entropy peak. This study demonstrates the potential of nanostructured Gd-Co multilayer stack for magnetic cooling applications. *Published by AIP Publishing.*

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I. INTRODUCTION

Recently, magnetic refrigeration based on the magneto-caloric effect (MCE) of magnetic materials possesses the potential to substitute conventional gas compression refrigeration, and has been a topic of great interest for minimizing environmental impact.¹ The basic requirement for magnetic refrigeration materials is a large isothermal magnetic entropy change ($-\Delta S_M$). However, several features are required for the application in magnetic refrigeration, such as the Curie temperature (T_C) in the vicinity of the working temperature, a large magnetic entropy change over the entire temperature range of range of the cycle, an almost zero magnetic hysteresis, a reasonable thermal conductivity coupled with large electrical resistance, and a large adiabatic temperature change.² Therefore, the relative cooling power (RCP) is indeed considered to be the most important factor for assessing the usefulness of a magnetic refrigerant material.³

During the last decade, MCE has been investigated in a very wide spectrum of materials that include elemental metals,⁴ rare earth intermetallics,⁵ manganese oxide materials,⁶ Mn alloys,⁷ lanthanides and Heusler compounds,⁸ bulk Gd alloys,⁹ and thin films.¹⁰ However, investigating MCE in materials at nanoscale remains a novel endeavor and an open challenge. Indeed, recent studies on $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ compounds in various forms, ranging from bulk and nanoparticles to thin films, showed that reducing the dimensionality of the LCMO compound tend to broaden and shift the paramagnetic-ferromagnetic transition to lower temperatures, while decreasing the M_s and the magnitude of $-\Delta S_M$.¹¹ Other studies on Gd-based structures demonstrated that the MCE also depends on the size and dimension.^{12,13} Indeed, it appears that the

combination of interfaces, defects, and lower size can explain the enhancement of MCE for low dimension samples. For instance, a maximum of entropy was observed in Gd/W multilayer.¹⁴ A more recent study by Lambert *et al.* showed that MCE can be tuned by changing the concentration of 100 nm-thick Gd-Tb alloys and multilayers.¹⁵ Moreover, a recent publication has shown that the $\text{Gd}_5\text{Si}_{2.7}\text{Ge}_{1.3}$ thin film exhibits a large magnetic entropy change ($-\Delta S_M = 8.83 \text{ J kg}^{-1} \text{ K}^{-1}$ for $-\Delta M = 50 \text{ kOe}$) and present a broader magnetic response in comparison with its bulk counterpart.¹⁶ In this paper, we report on the successful enhancement of the RCP value through the engineering of Gd-Co alloys based multilayer stack.

II. SAMPLE STRUCTURE

The studied sample is a 100 nm-thick Gd-Co alloys-based multilayer stack, namely $\text{Ta}(3 \text{ nm})/[\text{Gd}_{1-x}\text{Co}_x/\text{Gd}_{1-y}\text{Co}_y]/(100 \text{ nm})/\text{Ta}(3 \text{ nm})$. The studied sample is grown on a glass substrate at room temperature (RT) using sputtering deposition with base pressure lower than 10^{-7} Torr and an Ar pressure fixed at 3.0 mTorr. The composition gradients were achieved by adjusting the deposition rates from the two independent magnetron guns. Sputtering rates for Gd and Co targets at 50 W and 100 W were 0.719 Å/s and 0.456 Å/s , respectively, as determined by low-angle X-ray reflectivity measurements. The schematic of the sample structure is illustrated in Fig. 1. Indeed, the sample is made of four 25 nm-thick Gd-Co alloy layers, while the composition of each layer is different and varies linearly from $x = 0.44$ for the first layer (bottom layer) to $x = 0.60$ for the fifth layer (top layer). A 3 nm-thick Ta buffer layer was used, whereas another 3 nm-thick Ta capping layer was used to prevent sample oxidation. Magnetic

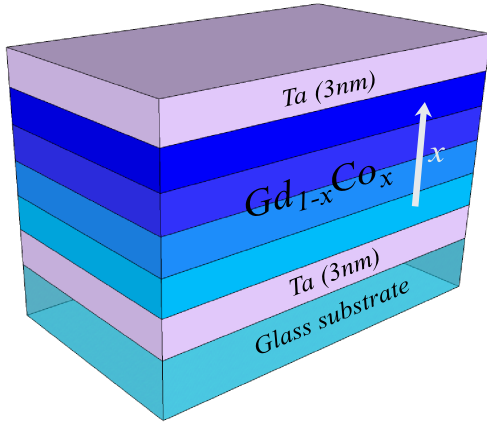


FIG. 1. Sketch of the studied Gd-Co alloys based multilayer stack. The sample is made of four 25 nm-thick $\text{Gd}_{1-x}\text{Co}_x$, where Co concentration x increases linearly from 0.44 for the first layer (bottom layer) to 0.60 for the fourth layer (top layer).

measurements were carried out by a Quantum Design SQUID Vibrating Sample Magnetometer.

III. RESULTS AND DISCUSSION

Figure 2 shows the temperature dependence of magnetization M under an applied magnetic field of 500 Oe for the studied Gd-Co-based multilayer stack sample. A typical paramagnetic to ferromagnetic phase transition can be observed at T_C . In order to estimate the T_C , we have used the inflection point method as described by Moreno-Ramirez *et al.*¹⁷ Following this, a value of $T_C = 230$ K can be determined from the minimum of the derivative of the magnetization $\frac{dM}{dT}$ as shown in the inset of Fig. 2.

The MCE in the studied Gd-Co-based multilayer stack was calculated from the isothermal magnetic entropy change ($-\Delta S_M$) deduced from the magnetization versus magnetic field measurements performed at different temperature below and above T_C and in magnetic fields up to 20 kOe. Figure 3 shows the temperature dependence of M obtained by sweeping the magnetic field from 0 to H_{max} using a sweeping speed

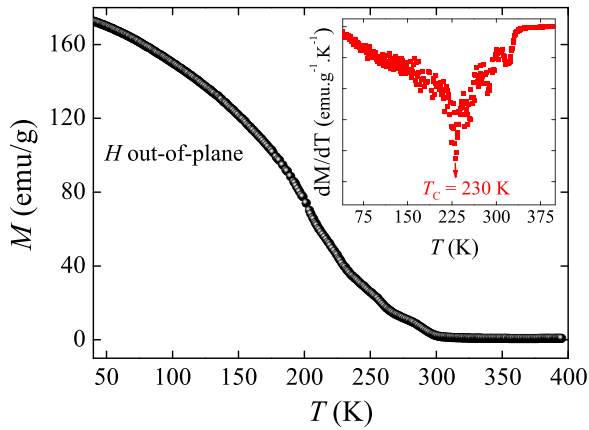


FIG. 2. Temperature dependence of the magnetization under a magnetic field of 500 Oe for the studied Gd-Co-based multilayer stack. The inset shows the evolution of $\frac{dM}{dT}$ as a function of temperature.

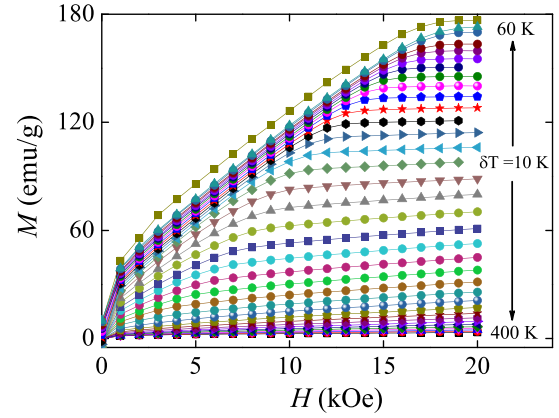


FIG. 3. Magnetic isotherms of the studied Gd-Co based multilayer studied as a function of the applied magnetic field from 0 to 20 kOe with a sweeping rate of 250 Oe/s.

of 250 Oe/s, with $H_{\text{max}} = 10, 15$ and 20 kOe, and for a temperature ranging from 60 to 400 K. The isothermal magnetic entropy change $-\Delta S_M$ is equal to $1.55 \text{ J kg}^{-1} \text{ K}^{-1}$ for a magnetic field swept from 0 to 20 kOe. Figure 4 shows the Arrott plots derived from $M(H)$ show a positive slope around T_C . According to the Banerjee criterion,¹⁸ this finding confirms that the measured paramagnetic-ferromagnetic transition is a second-order transition.

Furthermore, we compared the values of $-\Delta S_M$ obtained for the studied Gd-Co alloy-based multilayer stack with those obtained for a single 100 nm-thick Gd layer and for 100 nm-thick $\text{Gd}_{1-x}\text{Co}_x$ alloy layers with $x = 0.44, 0.48, 0.52$ and 0.56 (see Fig. 5). The values of $-\Delta S_M$ are obtained from the field dependence of the magnetization using the Maxwell relation

$$\Delta S_M(T, \Delta H) = \int_0^H \left(\frac{\partial M}{\partial T} \right)_H dH. \quad (1)$$

The maximum value of $-\Delta S_M$ is lower for the studied multilayer compared to the single Gd layer as well as the single Gd-Co alloy layers. However, the peak is much broader than for any of the single alloy layers. Note that the magnetic entropy is changed is not the only parameter to characterize the potential of a magnetic refrigerant. Indeed, the relative

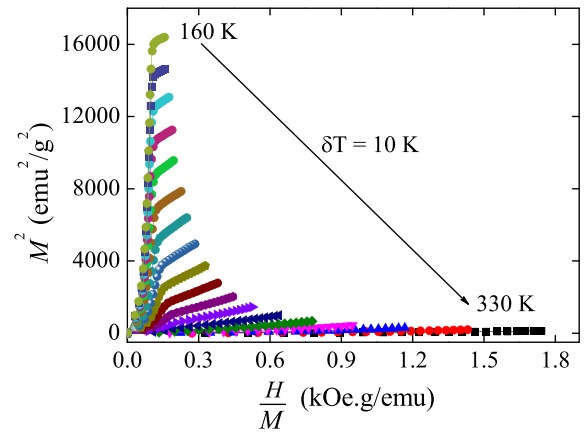


FIG. 4. Arrott plots showing the evolution of M^2 as a function of $\frac{H}{M}$ for the studied Gd-Co-based multilayer and for different values of temperature T .

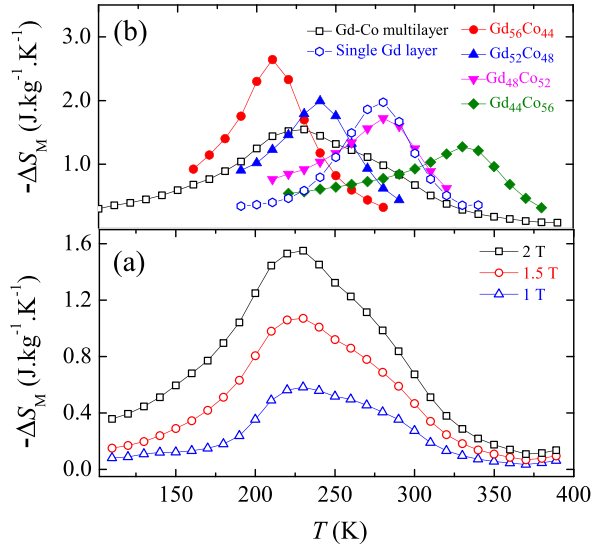


FIG. 5. Temperature dependence of the magnetic entropy change $-\Delta S_M$ (a) for the studied Gd-Co-based multilayer while sweeping the magnetic field from 0 to H_{Max} with $H_{\text{Max}} = 10$ kOe, 15 kOe and 20 kOe, (b) for the studied Gd-Co multilayer, a 100 nm-thick Gd layer, and four 100 nm-thick $\text{Gd}_{1-x}\text{Co}_x$ single alloy layers with $x = 0.44, 0.48, 0.52$ and 0.56 .

cooling power can also prove important.³ The RCP quantifies the amount of heat that can be transferred between the hot and the cold ends. For the point of view of applications, a large RCP over a wide temperature range coupled with a substantial MCE is desirable. The values of RCP can be calculated as

$$\text{RCP} = -\Delta S_M^{\text{max}} \delta T_{\text{FWHM}}, \quad (2)$$

with δT_{FWHM} is the full width at half maximum and $-\Delta S_M^{\text{max}}$ is the maximum of magnetic entropy change.¹⁹

From a technological point of view, it is important to obtain a large magnetic entropy change and a high RCP for a moderate magnetic field of 20 kOe. The measured RCP value for the Gd-Co alloys-based multilayer stack is 200 J/kg. This RCP value is slightly smaller than that reported for $\text{Gd}_5\text{Si}_2\text{Ge}_2$,⁹ FeRh polycrystalline alloy,²⁰ epitaxial MnAs,²¹ and $\text{Gd}_{48}\text{Co}_{52}$ amorphous ribbons²² for the same magnetic field change of 20 kOe. At the same time, the measured RCP value for the studied Gd-Co based multilayer is higher than that observed in manganite thin films, such as $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$,²³ $\text{La}_{0.67}\text{Ba}_{0.33}\text{Ti}_{0.02}\text{Mn}_{0.98}\text{O}_3$,²⁴ or $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ on SrRuO_3 super-lattices,²⁵ Gd/W multilayered films,²⁶ and $\text{Gd}_{62}\text{Co}_{38}$ amorphous ribbons.²⁷ We summarize in Table I the main performances related to the MCE of the above-presented materials as well as some others reported in the literature.

IV. CONCLUSION

In summary, we have engineered a Gd-Co-based multilayer stack sample to enhanced magneto-caloric properties. We have verified that the multilayer undergoes a second-order transition at the Curie temperature. The value of $-\Delta S_M$ and RCP for the studied Gd-Co alloys-based multilayer is compared to the one obtained for single Gd-Co alloy layers. The maximum $-\Delta S_M$ for the multilayer reach $1.54 \text{ J kg}^{-1} \text{ K}^{-1}$ for a magnetic field change from 0 to 20 kOe which is lower

TABLE I. Overview of the main performances related to the magneto-caloric effect for the studied Gd-Co alloys-based multilayer stack and for other materials reported from the literature.^{16,23–31}

Materials	$-\Delta S_M$ ($\text{J kg}^{-1} \text{ K}^{-1}$)	RCP (J/kg)	T_C (K)	ΔH (T)	References
Gd/W (40 nm)	1.41	52	275	1	26
$\text{Gd}_5\text{Si}_{2.7}\text{Ge}_{1.3}$ (788 nm)	3.7	75	190	2	16
Gd (30 nm)	1.6	70	285	1	28
MnAs (70 nm)	4	125	300	2	29
[Gd/Tb] (100 nm)	1.97	175	252	2	15
$\text{La}_{0.56}\text{Sr}_{0.44}\text{MnO}_3$ (31 nm)	0.3	10	70	1	30
$\text{La}_{0.67}\text{Ba}_{0.33}\text{Ti}_{0.02}\text{Mn}_{0.98}\text{O}_3$ (95 nm)	0.99	49	286	1	24
Ni-Mn-Ga (400 nm)	1.7	...	346	1	31
$[\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3/\text{SrTiO}_3]$ (20 nm)	1.1	15	321	1	23
$[\text{La}_{0.7}\text{Sr}_{0.33}\text{MnO}_3/\text{SrRuO}_3]$ (8 nm)	2.35	125	325	2	25
$\text{Gd}_{62}\text{Co}_{38}$ amorphous ribbons	2.8	81.4	193	1	27
$(\text{Gd}_{44}\text{Co}_{56})/(\text{Gd}_{48}\text{Co}_{52})/(\text{Gd}_{52}\text{Co}_{48})/(\text{Gd}_{56}\text{Co}_{44})$ (100 nm)	1.54	200	230	2	This paper

than for any of the single Gd-Co alloy layers. However, the $-\Delta S_M$ peak is much broader for the multilayer and the RCP value can reach 200 J/kg. Such enhancement of the RCP demonstrates the potential of Gd-Co based multilayers for magnetic refrigeration applications.

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