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Microstructure and Thermomagnetic Properties of As-Quenched $Gd_{75}Ge_{15}Si_5Pr_5$ Alloy

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Microstructure and thermomagnetic properties of the as-quenched $Gd_{75}Ge_{15}Si_5Pr_5$ (wt.%) alloy are studied. Multiphase composition of the investigated sample was confirmed by microstructural as well as magnetic investigations. The presence of the $Gd_{68}Ge_{18}Si_7Pr_7$ and $Gd_{76}Ge_{14}Si_5Pr_5$ regions with different atom concentration was confirmed by SEM/EDS analysis and temperature dependence of magnetic mass susceptibility. From DC magnetic measurements of magnetization versus temperature performed in zero-field/field cooled modes, and magnetic mass susceptibility investigations the Curie temperature (283 K and 278 K, respectively) and magnetic behavior were studied. Moreover, the structural investigations were performed as temperature dependence of specific heat capacity.

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1. Introduction

Gadolinium, lanthanum and other Rare Earth based materials are very interesting because show good magnetic properties and exhibit large magnetocaloric effect (MCE) [1–2]. Therefore, these alloys are intensively studied during the recent years [3–5]. Magnetic materials with the Curie point close to the room temperature are essential for potential applications in cooling technology [6–7]. MCE materials that are used in refrigeration promise energy saving as well as application of environmentally friendly technology.

In this paper, the microstructure, topography, structural changes and magnetic properties of the $Gd_{75}Ge_{15}Si_5Pr_5$ (wt.%) alloy are studied in wide range of temperatures and magnetic field.

2. Experimental procedure

As-quenched sample with a nominal composition of $Gd_{75}Ge_{15}Si_5Pr_5$ (wt.%) was prepared by an arc melter (Edmund Bühler GmbH) as ingot (about 10 g) on Cu water cooled plate under protective Ar atmosphere. Material was re-melted 10 times to guarantee homogeneous structure. Microstructure of the sample was analysed by Scanning Electron Microscopy equipped with Energy Dispersive X-ray Spectroscopy (SEM/EDS — Quanta 250, FEI). Temperature dependence of magnetization (in zero-field cooled (ZFC) and field cooled (FC) modes), DC hysteresis loops and AC magnetic mass susceptibility were recorded by VersaLab and Physical Properties Measurement Systems (Quantum Design). All magnetic investigations were performed in the temperature range 50–400 K and external magnetic field up to 3 T.

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3. Results and discussion

Figure 1 shows topography of the $Gd_{75}Ge_{15}Si_5Pr_5$ alloy observed by SEM equipped with secondary electron (SE) and backscattered electron (BSE) detectors. The SEM (SE) topography of the investigated alloy is typical for Gd-Ge-Si alloys. From BSE data it is seen that distribution of atoms in the investigated alloy is almost uniform, however some area with different atoms concentration were discovered in the analysed region. The results of chemical composition changes in the $Gd_{75}Ge_{15}Si_5Pr_5$ alloy performed with help of EDS detector are visible in Table 1. During the production process of the investigated material, the irregular regions with different content of Gd-Ge-Si-Pr were created. Two different regions with Gd-low ($Gd_{68}Ge_{18}Si_7Pr_7$) and Gd-high ($Gd_{76}Ge_{14}Si_5Pr_5$) content were observed. The volume fraction of these regions are 43% and 57%, respectively. However, from EDS mapping analysis it was seen that chemical composition of produced material was not changed during re-melting process.

TABLE I

Chemical composition of the regions in the $Gd_{75}Ge_{15}Si_5Pr_5$ alloy obtained from SEM/EDS investigations.

Sample [wt.%]	Chemical composition [wt.%]	Volume fraction [%]
As-quenched $Gd_{75}Ge_{15}Si_5Pr_5$	$Gd_{68}Ge_{18}Si_7Pr_7$	43
	$Gd_{76}Ge_{14}Si_5Pr_5$	57

It is well known that microstructure of the sample affects magnetic properties. Therefore, thermomagnetic properties of produced alloy were analysed in wide range of temperature and external magnetic field. Temperature behaviour of magnetization measured in ZFC and FC modes at the external magnetic field of 0.05, 0.1 and 1 T for the $Gd_{75}Ge_{15}Si_5Pr_5$ alloy is depicted in

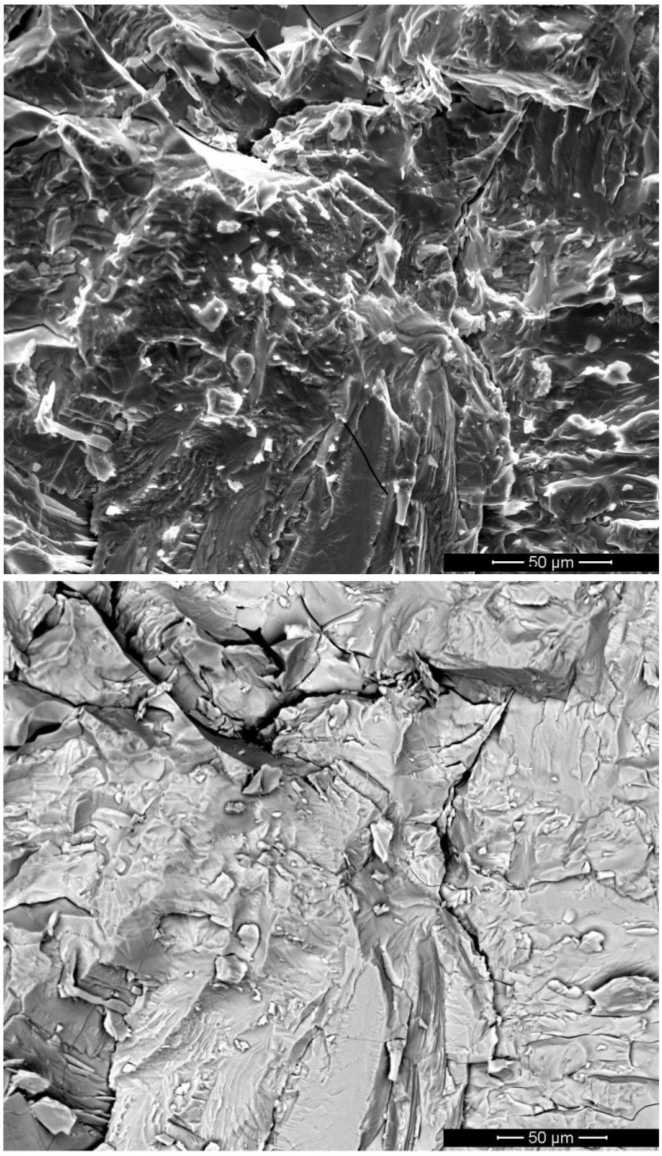

Fig. 1. SE (upper) and BSE (lower) SEM topographies of the as-quenched $Gd_{75}Ge_{15}Si_5Pr_5$ alloy.

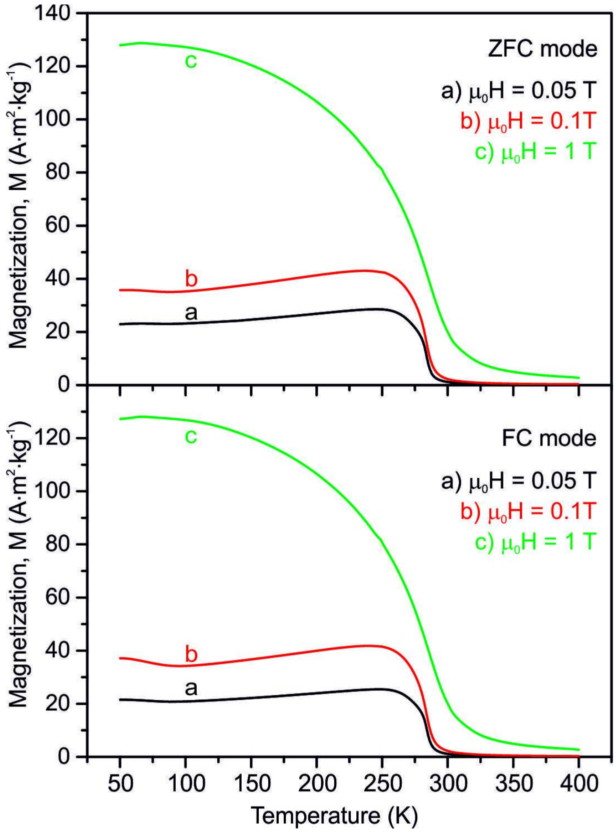

Fig. 2. Temperature dependence of magnetization for the $Gd_{75}Ge_{15}Si_5Pr_5$ alloy recorded in zero-field cooled (upper figure) and field cooled (lower figure) modes at the external magnetic field of 0.05, 0.1, and 1 T.

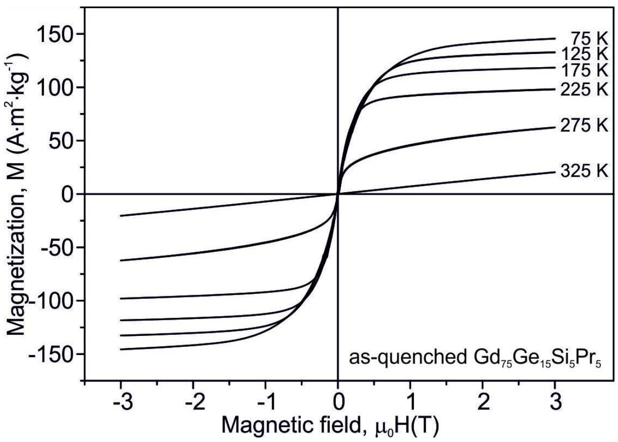

Fig. 3. DC hysteresis loops $M(H)$ for the as-quenched $Gd_{75}Ge_{15}Si_5Pr_5$ alloy measured at the indicated temperatures.

Fig. 1. It is seen that transformation from ferromagnetic to paramagnetic state close to the room temperature takes place. The Curie point T_C of produced material, calculated as derivative dM/dT performed for $M(T)$ curve taken at ZFC mode at external magnetic field of 0.05 T equals to 283 K, which is almost the same as for the $Gd_{75}Ge_{15}Si_5Ce_5$ alloy [8]. Above this temperature the material demonstrates paramagnetic behaviour. It is noteworthy that the magnetization curve recorded in ZFC and FC modes at the same external magnetic field shows identical character. Moreover, magnetization recorded at external magnetic field of 0.05 and 0.1 T slightly increases with temperature before the maximum value is reached and then suddenly drops down at the vicinity of T_C. For the sample measured at magnetic field $\mu_0 H = 1$ T, which is almost enough to saturate material, the typical $M(T)$ behavior characteristic for Heisenberg ferromagnets was observed.

Comparison of magnetic behavior of the as-quenched $Gd_{75}Ge_{15}Si_5Pr_5$ alloy at different temperatures (below and above the Curie point) by the help of hysteresis loops is shown in Fig. 3. As it can be seen from this figure the higher values of magnetization were measured at the lower values of temperature of measurement at the same external magnetic field. These results are in good agreement with data presented in Fig. 2. The coercive field for the investigated sample measured below the Curie point was less than 2.5 mT. Moreover, $M(H)$curve recorded for the investigated sample at temperature of 325 K shows a linear dependence, which is distinctive for paramagnetic materials.

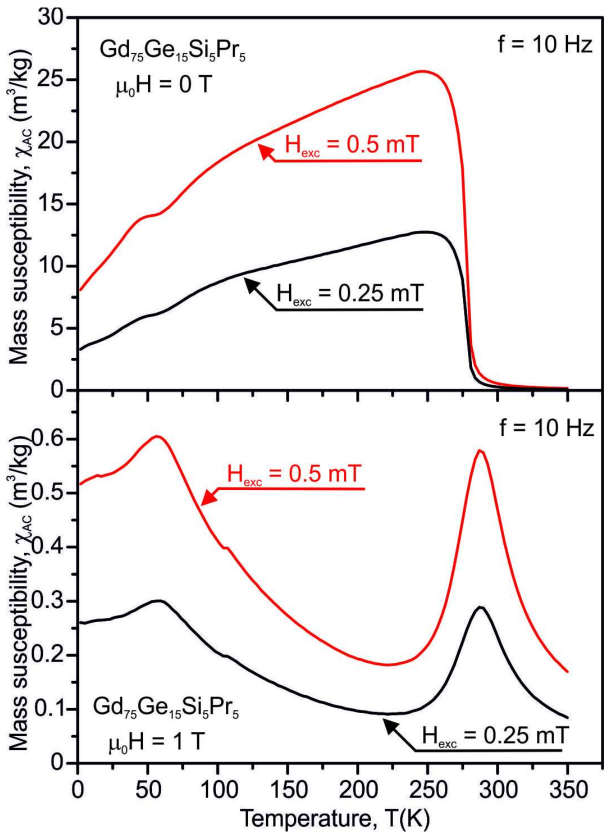

Fig. 4. Temperature dependence of AC magnetic mass susceptibility for the $Gd_{75}Ge_{15}Si_5Pr_5$ alloy traced for the same frequency of 10 Hz at zero field (upper figure) and external magnetic field $\mu_0 H = 1$ T (lower figure), with AC amplitude of excitation magnetic field of 0.25 mT and 0.5 mT.

To verify the Curie point and presence of magnetic regions with different atoms concentration, discovered by SEM/EDS, in the as-quenched $Gd_{75}Ge_{15}Si_5Pr_5$ alloy the most sensitive technique of AC magnetic mass susceptibility measurements were adopted. The temperature dependence of AC magnetic mass susceptibility (χ_{AC}) at different values of DC external magnetic field traced for excitation AC magnetic field (H_{exc}) of 0.25 and 0.5 mT is presented in Fig. 4. It is well seen that the investigated material shows different temperature behaviour of χ_{AC} at zero external magnetic field (Fig. 4 upper figure) and $\mu_0 H = 1$ T (lower figure), which is characteristic for non-single phase sample.

For the measurement performed at zero external magnetic field χ_{AC} increases with temperature to its higher value and then suddenly drops down in the vicinity of Curie point. Moreover, with increasing excitation AC magnetic field from 0.25 mT to 0.5 mT the higher values of AC magnetic mass susceptibility were measured for both zero and 1 T external magnetic field, as it is seen in Fig. 4. The Curie point calculated from $\chi_{AC}(T)$ for $H_{exc} = 0.25$ mT at zero field equals 278 K. The contribution of magnetic regions present in the investigated sample is well visible on Fig. 4 (lower figure). The additional low intensity component with maximum at 288 K is clearly seen during measurement at external magnetic field of 1 T. It is also worth noting, that for the investigated material AC magnetic mass susceptibility does not depend on frequency of measurement in the range 10 Hz–10 kHz.

Figure 5 shows temperature dependence of zero field specific sample heat capacity (H_C) related to 1 mol of Gd for the as-quenched $Gd_{75}Ge_{15}Si_5Pr_5$ alloy. The investigated sample shows a wide visible maximum in curve in the temperature range 220–274 K, related to two overlapped curves corresponding to $Gd_{68}Ge_{18}Si_7Pr_7$ and $Gd_{76}Ge_{14}Si_5Pr_5$ with different critical temperatures. The maximum value of the specific sample heat capacity recorded in the temperature 220–274 K equals to 76.93 J/(mol K) (at about 272 K). The structural transformation i.e. decreasing in H_C is visible at about 275 K, which is close to the magnetic transformation described by Curie point.

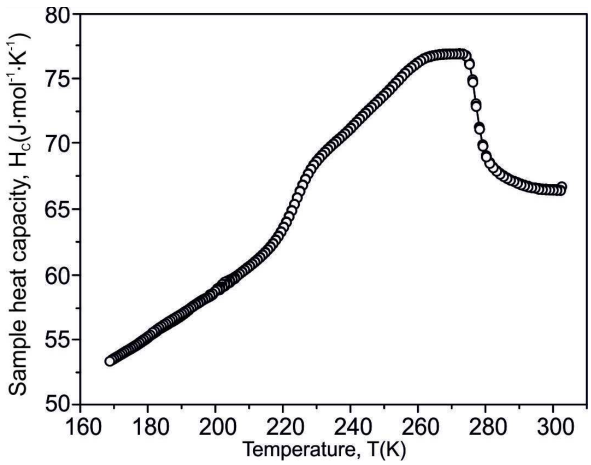

Fig. 5. Zero field specific sample heat capacity (H_C) related to 1 mol of Gd versus temperature for the as-quenched $Gd_{75}Ge_{15}Si_5Pr_5$ alloy.

4. Conclusions

The as-quenched $Gd_{75}Ge_{15}Si_5Pr_5$ (wt.%) alloy shows the presence of two different $Gd_{68}Ge_{18}Si_7Pr_7$ and $Gd_{76}Ge_{14}Si_5Pr_5$ regions. The Curie point calculated from temperature dependence of magnetization at external magnetic field of 0.05 T equals to 283 K, whereas T_C calculated from zero field AC magnetic mass susceptibility equals to 278 K. The difference in T_C results from the fact that both measurements were performed under different conditions i.e. $M(T)$ curve was recorded at external magnetic field of 0.05 T, whereas $\chi_{AC}(T)$ dependence was traced for H_{exc} = 0.25 mT at zero external magnetic field. Multiphase magnetic nature of the investigated material was also confirmed by additional peaks on $\chi_{AC}(T)$ curve traced at external magnetic field of 1 T. The zero field specific sample heat capacity shows wide spectrum with maximum at about 272 K.

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