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NEW MATERIALS OF RMn_2Ge_2 TYPE FOR THERMOMAGNETIC DEVICES

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The new type of the working media for thermomagnetic devices is proposed: the intermetallic compounds which have large magnetocrystalline anisotropy and exhibit the spontaneous phase transition from the antiferromagnetic (AF) to ferromagnetic (F) state when the temperature increases above the critical value T_t. Moreover, the anisotropy in AF state in the fields below the critical AF–F transition field must be much lower than in the F state. In the present paper the $Gd_{0.6}Sm_{0.4}Mn_2Ge_2$ compound belonging to the tetragonal RMn_2Ge_2 system is investigated. This compound has the critical temperature $T_t = 280$ K. The thermomagnetic cycle can be realized on the AF–F–AF transition near T_{t2}. The interval of the working temperatures can be shifted with the change of the Gd and Sm concentration.

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In the present paper we consider the new materials of the RMn_2Ge_2 type where R is a rare-earth metal as working media for thermomagnetic devices. These compounds crystallized in a tetragonal layered structure of the $ThCr_2Si_2$ type [1] (space group $I4/mmm$). The minimal distance between the same layers is equal to $c/2$ and the distance between two manganese atoms within the layer (d_{Mn-Mn}) is $a/\sqrt{2}$. The last parameter plays an important role in the explanation of the magnetic properties of these compounds. It is shown [2] that the exchange interaction between the Mn layers is ferromagnetic at $d_{Mn-Mn} > 0.286$ nm and antiferromagnetic in the contrary case.

$GdMn_2Ge_2$ is antiferromagnetic with $T_N = 365$ K [3]. Only Mn magnetic sublattice is ordered at high temperatures and at lower temperatures ($T < 95$ K) both Gd and Mn sublattices are ordered ferromagnetically but contrary to each other.

$SmMn_2Ge_2$ has a more complex magnetic behaviour [4]. Three spontaneous magnetic phase transitions were observed in this compound. At $T < T_{t1} = 64$ K both Sm and Mn magnetic moments are ordered parallel to each other. At higher temperatures only the manganese sublattice is ordered. Between T_{t1} and $T_{t2} = 196$ K this compound is antiferromagnetic, at $T_{t2} < T < T_c = 348$ K it has ferromagnetic order and above T_c it becomes paramagnetic. It was shown [5] that the $Sm_{1-x}Gd_xMn_2Ge_2$ powders exhibit the high temperature spontaneous AF $\rightarrow$ F

phase transition as well as the initial $SmMn_2Ge_2$ if $x < 0.65$. The critical temperature T_{t2} of this phase transition depends strongly on the composition and increases with the increase in Gd content. We choose $Gd_{0.6}Sm_{0.4}Mn_2Ge_2$ compound in which the critical temperature of AF $\rightarrow$ F transition is close to the room temperature ($T_{t2} = 280$ K).

The quasicrystals of a flat form were taken as the samples for the magnetic measurements. The thin single-crystal plates of the sample had common [001] direction but their basal planes were turned relatively to each other.

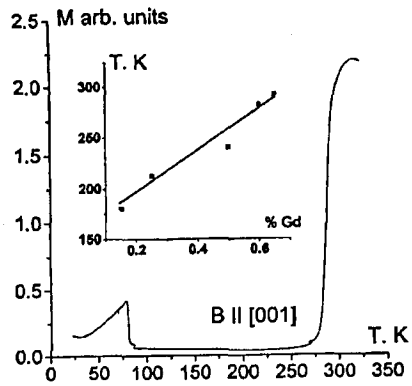

Fig. 1. Temperature dependence of the magnetization measured along [001] axis in the field of 0.1 T. The inset shows the concentration dependence of the critical temperatures taken from [5].

The temperature dependence of magnetization presented in Fig. 1 is similar to those observed for pure $SmMn_2Ge_2$ [4]. There are three temperature regions with different magnetic arrangement.

1) The very low magnetization value was observed at 4.2 K. Hence in the temperature region $T < T_{t1} = 81$ K it can be proposed that in $Gd_{0.6}Sm_{0.4}Mn_2Ge_2$ as well as in $GdMn_2Ge_2$, the Gd sublattice has the antiparallel orientation to Sm and Mn sublattices, although the existence of more complex magnetic structure cannot be ruled out.

2) In the temperature region 81 K $< T <$ 280 K the rare-earth sublattices are probably in the paramagnetic state and the Mn sublattice is in antiferromagnetic state.

3) The further increase in the temperature leads to the rearrangement of the Mn sublattice from AF to F state at $T_{t2} = 280$ K. This compound becomes paramagnetic at $T_c = 340$ K [5].

Magnetization curves measured along and perpendicular to the [001] axis are presented in Fig. 2. In the AF region (80 K$< T <$ 280 K) the external magnetic field $B \parallel c$ causes the magnetic phase transitions from the initial AF to F state at the critical value B_{cr}. In the case $B \perp c$ an angular structure appears. These field-induced phase transitions are accompanied by a small hysteresis. The interesting peculiarity of the observed magnetization curves is the equality of magnetic

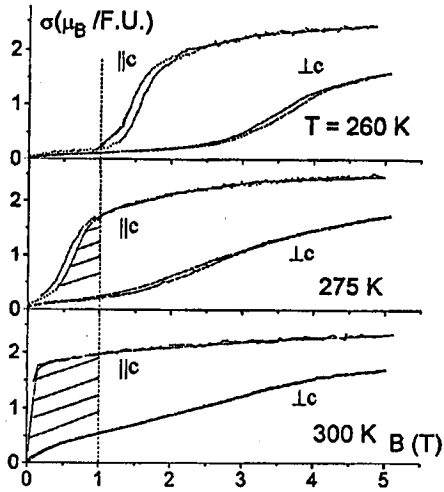

Fig. 2. Magnetization curves measured along and perpendicular to the [001] axis at various temperatures.

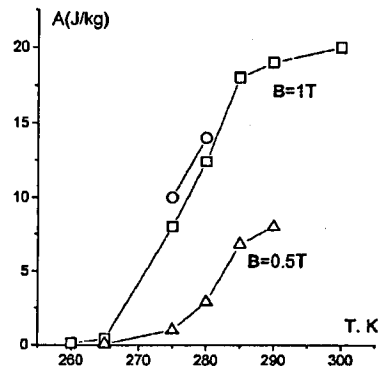

Fig. 3. The temperature dependence of the work connected with the rotation of the sample from the position $B \perp c$ to the position $B \parallel c$. Squares and circles correspond to the ascending and descending branches of hysteresis loop respectively.

susceptibilities in the easy and hard directions in low magnetic fields in the AF state. The latter was also observed in $GdMn_2Ge_2$.

In the F region (280 K< T < 340 K) the $Gd_{0.6}Sm_{0.4}Mn_2Ge_2$ compound has the relatively large anisotropy. The origin of anisotropy in these compounds is not clear. The anisotropy can be connected with Mn sublattice only, or rare-earth sublattice may also give its own contribution.

Such properties, i.e. an existence of the large anisotropy in F-region and the absence of the anisotropy in the AF-region in low fields can be used for the realization of the thermomagnetic cycle. At the initial position the magnetic field $B < B_{cr}$ is directed perpendicular to the [001] axis of the $Gd_{0.6}Sm_{0.4}Mn_2Ge_2$

sample at the temperature just below T_{t2}. The sample is heated, the temperature increases above T_{t2} and the sample undergoes to the F state with the large anisotropy. At this temperature the sample spontaneously rotates from the hard direction ($B \perp c$) to the easy direction ($B \parallel c$) doing useful work. Then the sample is cooled, undergoes to the AF state with the low anisotropy and must be returned in the initial position ($B \perp c$).

The work connected with the rotation of the sample in a constant magnetic field from the position ($B \perp c$) to the position $B \parallel c$ is equal to the area between the magnetization curves measured along hard and easy directions (shaded areas in Fig. 2). The temperature dependence of the work calculated by this way is presented in Fig. 3 for two applied fields — 1 T and 0.5 T. As it can be seen from Fig. 3 the useful work equal to 18 J/kg per cycle can be obtained in the field of 1 T when the initial temperature is equal to 285 K and the final temperature is equal to 265 K. The temperature interval of the cycle can be changed by changing Gd and Sm content (see the insert to Fig. 1).

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