Universal dependence of the magnetic entropy change versus temperature discovered earlier for magnetic materials with the second order transition is applied for the adiabatic temperature change versus temperature ($\Delta T(T)$) dependences. It is shown that the $\Delta T$ universal curve gives good description for the $\Delta T(T)$ experimental results obtained for gadolinium and TbCo$_2$ from the direct measurements in the vicinity of the Curie temperature.

1. INTRODUCTION

The main parameters, which characterize the magnetocaloric effect (MCE) are the adiabatic temperature change ($\Delta T$) and the isothermal magnetic entropy change ($\Delta S_m$). Both these parameters are important for application of a magnetic material in magnetic refrigerators, because the first determines the temperature span of the device and the second – its cooling capacity. In this connection intensive experimental studies of these parameters were carried out during the last decade (Tishin and Spichkin, 2003; Gschneidner et al. 2005; Pecharsky and Gschneidner, 1999, Bruck 2003).

The most experimental works are devoted to studies of magnetic field and temperature dependences of the isothermal magnetic entropy change $\Delta S_m$, because it is important from the practical point of view and can be rather easy determined from magnetization isotherms measurements with the help of Maxwell’s relation. The adiabatic temperature change $\Delta T$ measurements require more complex methods – this parameter can be determined from the heat capacity data obtained in zero and nonzero magnetic fields and also from direct measurements of the sample temperature change under application of the magnetic field. The most interesting temperature range of the MCE investigations is the vicinity of the Curie temperature ($T_C$), because here the effect reaches its maximum values and magnetic materials selected for application as working bodies in magnetic refrigerators are used just near their $T_C$. It should be noted that $\Delta S_m$ and $\Delta T$ measurements usually require vast time and essential efforts.

Franco et al. (2006) studied theoretically the field dependence of the isothermal magnetic entropy change of materials with a second order phase transition near the Curie temperature and showed that the $\Delta S_m(T)$ curves measured with different applied fields should collapse into a single universal curve in rescaled $\Delta T$ and $T$ axis. The collapse of $\Delta S_m(T)$ curves determined for different $H$ for the single curve was demonstrated for Fe based amorphous alloys (Franco et al. 2006) and for crystalline materials as Gd and (Er$_x$Dy$_{1-x}$)Al$_2$ (Franco et al. 2007).

Until now the universal curves method was applied to the isothermal entropy change $\Delta S_m$. The aim of this work is to show its applicability to the adiabatic temperature change $\Delta T$.

2. EXPERIMENT

To illustrate the applicability of the proposed method two set of data have been used: first- experimental data obtained in this work for classical ferromagnet Gd and second - experimental data from the work...
Spichkin et al. (2009a) for intermetallic compounds TbCo₂. Polycrystalline Gd (99.5 at % pure) was obtained from a commercial source. Polycrystalline TbCo₂ sample used in the work Spichkin et al. (2009a) was prepared by arc-melting in the Material Preparation Center of the Ames Laboratory. Terbium was 99.5+ at.% pure with the major impurities being the interstitial elements (O, C, F, and N). Cobalt was obtained from commercial source and were 99.9 wt.% pure. X-ray powder diffraction analysis confirmed that TbCo₂ sample was single phase.

The method of the direct $\Delta T(H)$ measurements was described in detail by Tishin et al. (2007) and Spichkin et al. (2009b). The measurements were made on the Magnetocaloric Measuring Setup “MagEq MMS 801” manufactured by Advanced Magnetic Technologies and Consulting, Ltd., Moscow, Russia. The magnetic field was created by permanent magnet Halbach magnetic field source with changeable magnetic field in its working bore. $\Delta T$ values were measured by a differential thermocouple with the measuring junction clamped between two pieces of the material under investigation and a reference junction placed on the non-magnetic metallic sample holder near the sample. $\Delta T$ and $H$ values were recorded simultaneously and continuously over the whole cycle of the magnetic field change, which makes it possible to construct the $\Delta T(H)$ curves and on the basis of this data get $\Delta T(T)$ at necessary magnetic field change ($\Delta H$) value. The measurements were made near the corresponding Curie temperatures $T_C$ at the magnetic field change rate of 1 T/s. The maximum magnetic field ($H$) was 1.87 T.

3. RESULTS AND DISCUSSION

The field dependence of the isothermal magnetic entropy change of materials with a second order phase transition can be expressed as a potential law:

$$\Delta S_M \sim H^n.$$  

The exponent $n$ at the Curie temperature corresponds to (Franco et al. 2006):

$$n = 1 + \frac{\beta - 1}{\beta + \gamma} = 1 + \frac{1}{\delta} \left( 1 - \frac{1}{\beta} \right),$$  

(2)

$\gamma$ and $\beta$ are critical exponents of the material and critical exponent $\delta$ is determined by the relation:

$$\beta \delta = \beta + \gamma.$$  

(3)

It was established that there are three distinct temperature regions regarding $n$:

- $n = 1$ well below $T_C$,
- $n = 2$ well above $T_C$,
- and the intermediate region in the vicinity of $T_C$, where $n$ presents a minimum.

Experimental investigations of the critical exponents $n$, $\gamma$, and $\beta$ of amorphous alloy Fe₈₃Zr₆B₁₀Cu₁ made on the basis of magnetization and $\Delta S_M$ data showed remarkable agreement with the proposed model.

Using the conclusion about the general character of the $\Delta S_M$ on $H$ dependence it was suggested that the $\Delta S_M(T)$ curves measured with different applied fields should collapse into a single universal curve when properly rescaled. The $\Delta S_M$ axis was rescaled by normalization with the respective $\Delta S_M$ peak value:

$$\Delta S_M'(T, H) = \Delta S_M(T, H) / \Delta S_M^{pk}(H),$$  

(4)

where $\Delta S'$ is the rescaled axis, and $\Delta S^{pk}$ is the peak value for the correspondent applied magnetic field. The temperature axis was rescaled in a different way below and above $T_C$, by imposing that the position of two additional reference points in the rescaled curve $\theta$ correspond to $\theta = \pm 1$:

$$\theta = \begin{cases} 
-\frac{(T - T_C)}{(T_1 - T)}, & T \leq T_C \\
\frac{(T - T_C)}{(T_2 - T)}, & T > T_C 
\end{cases},$$  

(5)
where $T_{r1}$ and $T_{r2}$ are the temperatures of the two reference points that was selected as those corresponding to $0.5\Delta S_M$. The collapse of $\Delta S_M(T)$ curves determined for different $H$ for the single curve using the described procedure was demonstrated for Fe$_{83}$Zr$_6$B$_{10}$Cu$_1$ amorphous alloy (Franco et al. 2006). Later Franco et al. (2008) showed that the rescaling procedure can be simplified by using only one reference temperature $T_r$ and either of the expressions in eq. (5). $\Delta S_M$ axis in this case is rescaled by the $\Delta S_M$ value at this reference temperature ($\Delta S_M(T_r, H)$).

As it is known $\Delta S_M$ and $\Delta T$ are the values interrelated via the equation (Tishin and Spichkin 2003):

$$\Delta S_M(T, H) = -\frac{C_H}{T} \Delta T(T, H),$$

where $C_H$ is the heat capacity. Because of that it can be proposed that the adiabatic temperature change $\Delta T(T)$ dependences measured at various magnetic fields can also collapse to the $\Delta T$ universal curve analogous to the $\Delta S_M$ universal curve with the help of the procedure described above.

We measured $\Delta T(H)$ curves of polycrystalline Gd and intermetallic compound TbCo$_2$, which undergo the second order magnetic phase transition into ferromagnetic at the Curie temperature $T_C$ (Taylor et al. 1972). The Curie temperatures of Gd and TbCo$_2$ were determined earlier to be 294 and 231 K, respectively (Dankov et al. 1998; Tishin and Spichkin 2003). Fig. 1 shows adiabatic temperature change versus temperature curves for Gd and TbCo$_2$ determined from experimental $\Delta T(H)$ curves measured at the magnetic field change rate of 1 T/s under increasing of the magnetic field. The specific adiabatic temperature change $\Delta T/\Delta H$ values determined for Gd and TbCo$_2$ from the experimental data were 2.4 and 1 K/T, correspondingly, which is in good agreement with previous literature results (Tishin and Spichkin 2003).

Figure 1. Adiabatic temperature change temperature dependences for Gd and TbCo$_2$ determined from experimental $\Delta T(H)$ curves measured with the magnetic field change rate 1 T/s. The upper curves are for the magnetic field 1.87 T and the lowest curves are for the magnetic field 0.758 T and 0.697 for Gd and TbCo$_2$, correspondingly. The curves are shown with the magnetic field step of about 0.05 T.
Figure 2. Normalized $\Delta T(T)$ curves for Gd for $H = 1.87, 1.867, 1.851, 1.841, 1.829, 1.816, 1.802, 1.786, 1.749, 1.729, 1.707, 1.684, 1.659, 1.634, 1.607, 1.579, 1.549, 1.519, 1.487, 1.454, 1.419, 1.385, 1.349, 1.312, 1.274, 1.234, 1.195, 1.154, 1.114, 1.072, 1.028, 0.984, 0.939, 0.894, 0.849, 0.805, 0.758 T.

Figure 3. Normalized $\Delta T(T)$ curves for TbCo$_2$ for $H = 1.870, 1.867, 1.862, 1.854, 1.845, 1.834, 1.820, 1.805, 1.787, 1.767, 1.746, 1.722, 1.697, 1.670, 1.642, 1.612, 1.579, 1.545, 1.509, 1.472, 1.434, 1.393, 1.351, 1.308, 1.263, 1.216, 1.169, 1.121, 1.071, 1.020, 0.968, 0.915, 0.862, 0.807, 0.752, 0.697 T.

On the basis of these dependences $\Delta T(T)$ curves in the normalized $\Delta T$ and $T$ axis were plotted following the procedure from the work of Franco et al. (2008) with one reference point – Figs. 2 and 3. As one can see from Fig. 2 and 3, there is a rather good collapse of the experimental normalized $\Delta T(T)$ curves into one universal curve in the paramagnetic region and in the magnetically ordered state near the Curie temperature. In low temperature region there is some divergence of the curves for the magnetic fields less than 1 T. It can be related with demagnetization field effects, which are rather pronounced in the temperature region with high magnetization (Caballero-Flores et al. 2009).

4. CONCLUSIONS

In conclusion, the normalized $\Delta T(T)$ curves were constructed for polycrystalline Gd and TbCo$_2$ samples with the second order magnetic phase transition from the experimental $\Delta T(H)$ curves measured by the direct method. It was shown that the universal scaling dependence of the magnetic entropy change versus temperature discovered earlier for magnetic materials with the second order transition (Franco et al. 2006) is also valid for the adiabatic temperature change versus temperature curves in the paramagnetic state and in vicinity of the Curie temperature.
ACKNOWLEDGEMENTS

Y.I. Spichkin and I. Zubkov thank Advanced Magnetic Technologies and Consulting Ltd. for financial support of this work. The authors thank K.A. Gschneidner, Jr. and V.K. Pecharsky for the possibility to use the experimental data for TbCo₂ sample obtained in the work of Spichkin et al. (2009a) at low magnetic field change rate for numerical evaluations made in this work. V. Franco acknowledges the financial support of the Spanish Ministry of Science and Innovation and EU FEDER (Project MAT 2007-65227 and CIT-420000-2008-9), and the PAI of the Regional Government of Andalucía (Project P06-FQM-01823).

REFERENCES


