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Magnetic properties and magnetocaloric effect in TmZnAl and TmAgAl compounds

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ABSTRACT

The magnetic and magnetocaloric properties in the equiatomic intermetallic compounds of TmZnAl and TmAgAl have been investigated. The compounds undergo a second order magnetic transition from paramagnetic to ferromagnetic state around its own Curie temperatures T_C ~2.8 K and 3.3 K for TmZnAl and TmAgAl, respectively. A considerable reversible magnetocaloric effect (MCE) was observed at low temperature. For a magnetic field change of 7 T, the maximum values of magnetic entropy change ($-\Delta S_M^{max}$) and relative cooling power (*RCP*) are 11.8 J/kg K and 289 J/kg for TmZnAl, respectively, and the corresponding values for TmAgAl are 14.1 J/kg K and 315 J/kg.

1. Introduction

The technique of magnetic refrigeration based on the magnetocaloric effect (MCE) has attracted much interesting due to its energy efficiency and environmental-friendly features over to the common gas-compression refrigeration [1–6]. The MCE is a magneto thermodynamic phenomenon, which manifests as an adiabatic temperature (ΔS_M) or an isothermal magnetic entropy (ΔT_{ad}) when the material is exposed to a varying magnetic field. A large value of MCE is considered to be the most important requirement of the application, and therefore it is desirable to be found new materials with a large MCE at low magnetic fields, a wide temperature range and a small thermo-magnetic hysteresis. For this purpose, the magnetic properties and MCE in lots of rare-earth based compounds have been systematically investigated, and some of them are found to possess promising MCE properties, such as ferromagnetic TmCuAl [7], HoAgGa [8], Eu_4PdMg [9] and $ErMn_2Si_2$ [10] etc., metamagnetic Gd_3Ru [11] and TmZn [12] etc., magnetic superconductors $RENi_2B_2C$ [13,14], as well as HoPdIn [15] and $TbMn_2Si_2$ [16], which have two successive magnetic transitions.

The ternary intermetallic compounds of *RET*Al (*RE* = rare earth, and *T* = transition metal) have attracted much attentions owing to their interest chemical and physical properties [7,12,15,17–24]. The *RET*Al show a very rich variety of crystallographic structures depending on the constituent element and composition. Zhang et al. have investigated the MCE in the *RE*CoAl (*RE* = Gd, Tb, Dy and Ho) compounds, and found that *RE*CoAl could be candidate materials for magnetic refrigeration in the temperature range 10–100 K [17]. Large magnetic entropy change and refrigerant capacity have been observed in *RE*CuAl (*RE* = Gd, Dy, Ho and Er) compounds [18,19]. Recently, the magnetic and magnetocaloric properties in *RE*FeAl (*RE* = Gd, Tb, Dy Ho and Er) have been investigated systematically. It was found that *RE*FeAl exist a wide working temperature range from 55 K to 265 K, which is benefit to design a composite material used as a magnetic refrigerant [20–22]. We have studied the magnetic and magnetocaloric effect in *RE*AgAl (*RE* = Er and Ho), and the values of $-\Delta S_M^{max}$ were evaluated to be 10.5 and 10.3 J/kg K for a magnetic field change of 0–5 T for *RE* = Er and Ho, respectively [23]. Very recently, Mo et al. investigated the MCE in several Tm-based intermetallic compounds belonging to *RET*Al series, and a low-field giant MCE was found in TmCuAl and TmCoAl which is benefit for application [7,24]. In the present study, we have successfully synthesized two Tm-based ternary equiatomic compounds of TmZnAl and TmAgAl, and investigated the magnetic and magnetocaloric properties. A considerable reversible MCE in

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both compounds has been observed, which can provide some new data and valuable information for searching new magnetic materials at low temperature magnetic refrigeration.

2. Experimental

The TmZnAl and TmAgAl compounds in the polycrystalline form were prepared by the induction melting method. The starting high purity element (all better than 99.9%) of Tm, Zn, Ag and Al were weighted and arc-welded in a tantalum tube under an argon pressure of ca. 80 kPa. Then, the tantalum crucible was placed in a water-cooled sample chamber of an induction furnace and heated up to 1273 K for 4 min, following by 3 h annealing at 873 K. Both samples were confirmed to be single phase by X-ray diffraction (XRD) using Rigaku RINT 2200 diffractometer. The magnetic measurements were carried out by using a commercial vibrating sample magnetometer (VSM) which is an option of the physical property measurement system (PPMS-9, Quantum Design).

3. Results and discussion

The room temperature X-ray powder diffraction experimental (XRD) patterns for TmZnAl and TmAgAl compounds are revealed in Fig. 1. The XRD data confirmed that both compounds are crystallized in a single phase. The lattice parameters a and c were estimated to be 4.467 and 6.943 Å for TmZnAl; and to be 4.533 and 7.850 Å for TmAgAl, respectively. Fig. 2(a) and (b) show the temperature dependence of the zero field cooling (ZFC) and field cooling (FC) magnetization (M) under the applied magnetic field of 0.2 T for TmZnAl and TmAgAl, respectively. A distinct point of inflexion in magnetization curves is observed at ~2.8 and 3.3 K for TmZnAl and TmAgAl, respectively, which signifies a paramagnetic to ferromagnetic (PM-FM) transition. It is well known that the magnetism of intermetallic compounds of rare earths and non-magnetic metals is mainly originated from the rare earth sublattice, *i. e.* Tm–Tm sbulattice for present compounds. The difference of T_C for TmZnAl and TmAgAl is probably related to difference of the ion radius of Zn and Ag which will change the distance of Tm–Tm sublattice, then affect the exchange constant. Additionally, no obvious thermal hysteresis can be observed between ZFC and FC

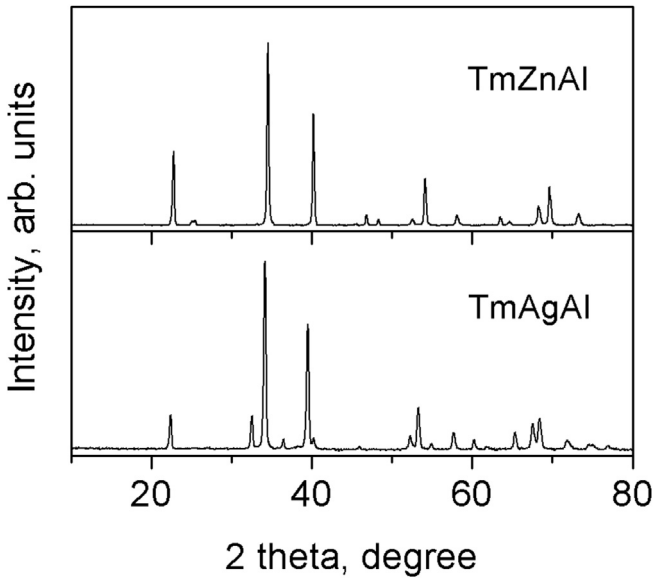

Fig. 1. The room temperature XRD patterns for TmZnAl and TmAgAl compounds.

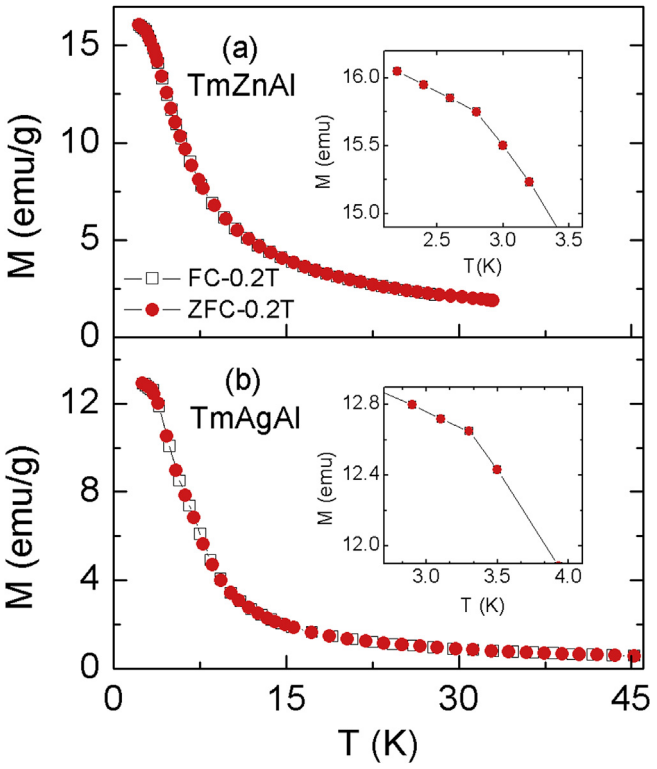

Fig. 2. Temperature dependence zero-field cooling (ZFC) and field cooling (FC) magnetization (M) under the magnetic fields of 0.2 T for TmZnAl (a) and TmAgAl (b) compounds, respectively. Insets of (a) and (b) show an expand view near the phase transition for TmZnAl and TmAgAl, respectively.

M-T curves which is benefit for application. The temperature dependence of the magnetization M (left side) and the reciprocal susceptibility $1/\chi$ (right side) for TmZnAl and TmAgAl under a high magnetic field of 1 T are shown in Fig. 3(a) and (b), respectively. We can observe that M increases continuously with decreasing temperature in both compounds. The high temperature reciprocal susceptibility for both compounds follows the Curie–Weiss law, $1/\chi = (T-\theta_p)/C$, where C is the curie constant and θ_p is the paramagnetic Curie constant. From a linear fitting of the high temperature data, the effective magnetic moment (μ_{eff}) is obtained to be 7.64 and 7.71 μ_B for TmZnAl and TmAgAl, respectively, which is close to that of the free ion value of Tm^{3+} (7.56 μ_B). The corresponding θ_P is evaluated to be 8.1 and 4.3 K, respectively. The positive values of θ_P further confirm the ferromagnetic ground state for both compounds.

To evaluate the MCE of TmZnAl and TmAgAl, a set of magnetic isothermals with increasing and decreasing field were measured for both compounds up to 7 T. Selected and representative $M(H)$ curves are shown in 4 (a) and 5 (a) for TmZnAl and TmAgAl, respectively. It is well known that the MCE has a strong correlation with the order of the corresponding magnetic phase transition, it is important to understand the nature of magnetic transition in TmZnAl and TmAgAl compounds. According to Banerjee criterion [25], the signal of the slope of H/M versus M^2 can determine the nature of the magnetic phase transition, with a positive slope corresponding to a second order transition and a negative slope at some point corresponding to a first order transition. Thus, the Arrott plots, H/M versus M^2, have been constructed based on the M-H data, and shown in Figs. 4(b) and 5(b) for TmZnAl and TmAgAl, respectively. By this criterion, neither the inflection point nor negative slopes can be observed, indicating TmZnAl and TmAgAl