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Investigations on magnetic and magnetocaloric properties of the intermetallic compound TbAgAl

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Magnetic and magnetocaloric properties of the intermetallic compound TbAgAl have been investigated. Temperature dependence of magnetization data revealed that TbAgAl exhibits magnetic ordering at 59 K and possesses competing ferromagnetic-antiferromagnetic interactions, which leads to the formation of Griffiths-like phase. The field dependence of magnetization data shows the existence of a metamagnetic-like transition at a critical field of 10 kOe. Unusually potent relaxation effects are seen in the time dependence of magnetization data in the ordered phase. The presence of spin-glass-like state is found to affect magnetocaloric properties of this compound.

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I. INTRODUCTION

A rich variety of magnetism exhibited by rare earth based intermetallic compounds make them suitable candidate for fundamental studies, as well as for applications based on magnetostriction, magnetoresistance, and magnetocaloric effect (MCE).1–6 MCE is defined as the thermal response of a magnetic material to an applied field and manifests itself as adiabatic temperature change ($\Delta T_{\text{ad}}$) or isothermal magnetic entropy change ($\Delta S_{\text{m}}$) when the material is subjected to a varying magnetic field. It is over a decade now since the first observation of giant MCE, resulting from the concomitant magnetic and structural transitions in the intermetallic compound Gd$_2$Si$_2$Ge$_2$ near its ferromagnetic transition was reported.7 Since then, there has been a tremendous activity in the field of MCE in general and on the study of magnetothermal properties of materials with coupled magnetic and structural transitions in particular.5,6,8–14 The experimental investigations have revealed that apart from the compounds having magnetostructural transitions, alloys having multiple magnetic transitions also possess enhanced magnetocaloric properties.15–17 Owing to the presence of the large number of soft modes, magnetically frustrated systems have also been reported to exhibit considerable MCE.5,18,19 It is well known that in any magnetic material the magnetic frustration is an essential ingredient for the formation of spin glasslike state20 and, therefore, materials exhibiting glassy features are also potential candidates to probe the correlation between magnetism and MCE behavior. Suresh et al.21 reported that in ternary RAgAl compounds the spin-glass-like state coexists with long range order. In this context, we have carried out further investigations on the magnetic and magnetocaloric properties of TbAgAl and results are presented in this manuscript. The magnetization data reveal that the title compound possesses competing ferromagnetic and antiferromagnetic interactions. The presence of contending interactions leads to the formation of Griffiths-like phase in the paramagnetic regime. The occurrence of considerable MCE over a wide temperature range renders TbAgAl as potential candidate for refrigeration applications around 60 K.

II. EXPERIMENTAL DETAILS

The TbAgAl alloy, weighing 2 g, was prepared by arc melting of stoichiometric amounts of the high purity (>99.9 wt %) constituent elements on a water cooled copper hearth, under argon atmosphere. To ensure the homogeneity, the resulting ingot was flipped upside down and remelted six times and was annealed for 1 week at 600 °C. The phase purity of the sample was examined by the Rietveld refinement to the experimental data. The refinement of the intermetallic compound TbAgAl obtained using the Cu Kα radiation and the time dependence of magnetization was measured immediately after the field was turned off.

III. RESULTS AND DISCUSSION

Figure 1 shows the room temperature x-ray diffraction pattern of TbAgAl obtained using the Cu Kα radiation and Rietveld refinement to the experimental data. The refinement reveals that the compound is single phase with the CeCu$_2$-type orthorhombic structure. The lattice parameters $a$, $b$, and $c$ are 4.5878(2), 7.1649(4), and 7.8643(4) Å, respectively. The lattice parameters obtained from the fit are found...
to be in good agreement with the previous report. The refinement yields a density of 10.981 g/cm³ for this compound.

Temperature \((T)\) dependencies of magnetization \((M)\) data have been collected in zero-field-cooled (ZFC) and field-cooled (FC) protocols under various applied magnetic fields \((H)\) and the representative ZFC-FC \(M(T)\) data obtained in \(H=5\) kOe are shown in Fig. 2. The \(M(T)\) plots at 100 Oe have been shown as an inset in Fig. 2. The low field data reveal that TbAgAl exhibits ferrimagnetic-like ordering with an ordering temperature \((T_C)\) of 59 K, which compares well with the previous report. We note that the ZFC-FC \(M(T)\) data show thermomagnetic irreversibility and, for \(H=100\) Oe, the bifurcation between the ZFC and FC \(M(T)\) data starts at \(T_C\). With increasing field the divergence between the ZFC-FC \(M(T)\) is found to start at temperatures below \(T_C\) and, for \(H=5\) kOe, the bifurcation begins at 34 K. It is well known that the thermomagnetic irreversibility between the ZFC and FC \(M(T)\) data is a typical feature associated with spin glass system; however, it may also be observed in ferromagnetic materials having large anisotropy, in materials with coexisting ferromagnetic and antiferromagnetic interactions, and in antiferromagnets having random orientation of crystallites. In view of the low \(T_C\) and considerably large magnetocrystalline anisotropy associated with the rare earth sublattice, the thermomagnetic irreversibility in the title compound may partly be attributed to domain wall pinning effect.

Figure 3 shows the temperature dependencies of the inverse susceptibility \((H/M)\) of TbAgAl and the Curie–Weiss \((M/H=C(T−\theta_P))\); where \(C\) and \(\theta_P\) are the Curie constant and the paramagnetic Curie temperature, respectively) fit to it. We note that above 90 K, the inverse susceptibility obeys the Curie–Weiss law with an effective moment \((\mu_{eff})\) and paramagnetic Curie temperature \((\theta_P)\) of 9.75\(\mu_B/\text{Tb}^{3+}\) and 43 K, respectively. The \(\mu_{eff}\) determined from the fit is found to be in good agreement with the \(\text{Tb}^{3+}\) free ion value of 9.72\(\mu_B\). It may be mentioned here that the \(\theta_P\) represents the sum of all the magnetic interactions present in system. Thus, observation of positive \(\theta_P\) reflects that the TbAgAl predominantly possesses ferromagnetic interaction; however, the lower value of \(\theta_P\) as compared to that of \(T_C\) indicates the presence of competing ferromagnetic and antiferromagnetic interactions in this compound. At this juncture, it is worth mentioning that the presence of competing interactions can also cause the occurrence of thermomagnetic irreversibility and, therefore, the large difference seen between the ZFC-FC \(M(T)\) data of TbAgAl may originate from competing interactions as well.

It may be seen from Fig. 3 that above 90 K the inverse susceptibility obeys the Curie–Weiss behavior; however, between 90 K and \(T_C\) it exhibits a negative deviation from the Curie–Weiss behavior. Furthermore, with increasing \(H\), the extent of deviation is found to decrease (see Fig. 3). The negative deviation of \(H/M\) from the Curie–Weiss behavior is considered as hallmark of the Griffiths-like phase. A Griffiths-like phase is defined as an intermediate magnetic state between true paramagnetic state and magnetically ordered state where the magnetization ceases to be an analytic function of field when field approaches zero and is usually observed in materials having competing magnetic interactions. The presence of competing interactions leads to formation of dilute ferromagnetic clusters in the paramag-
netic matrix, which results in the negative deviation of $H/M$ from Curie–Weiss law in low fields. However, in the presence of high fields the contribution to magnetization from the paramagnetic matrix increases, which leads to the reduction in the extent of deviation.\textsuperscript{28}

Figure 4 shows the field dependencies of magnetization data of TbAgAl collected at various temperatures. $M(H)$ isotherms were obtained after reaching the desired temperature under ZFC condition. At 4 K the low field magnetization varies linearly with field and at about 10 kOe the $M(H)$ data show signature of a metamagnetic-like transition [see Fig. 4(a)]. We note that although the metamagnetic transition leads to a rather faster increase in the magnetization, the high field $M(H)$ data exhibit lack of saturation. At $T=4$ K and $H=80$ kOe, the magnetization is found to be $5.7 \mu_B/\text{Tb}^{3+}$, which is significantly lower than the theoretically expected (gJ) value of $9 \mu_B$ for Tb$^{3+}$ ion. The nonsaturation tendency is seen in the $M(H)$ isotherms collected at high temperatures as well [see Fig. 4(b)]. It may be mentioned here that the nonsaturation in magnetization is generally observed in antiferromagnets, in ferromagnets having significant antiferromagnetic component, or in compounds with large magnetocrystalline anisotropy.\textsuperscript{20–22,25,26} However, a nonsaturation tendency in the field dependence of magnetization data may also be observed in spin glasses or intermetallic compounds having inhomogeneous magnetic state.\textsuperscript{20,21}

In order to further probe the low temperature magnetic state of TbAgAl, we have also performed time dependent magnetization measurements and results are shown in Fig. 5. It may be seen from the figure that the time ($t$) dependence of isothermal remanent magnetization ($M_{IRM}$) data show a strong time dependence. The analysis of the data shows that $M_{IRM}$ versus $t$ has logarithmic dependence of the form $M_{IRM}(t)=M_{IRM}(0)-S \ln(1+t)$, where $M_{IRM}(0)$ and $S$ are initial zero field remanent magnetization and magnetic viscosity, respectively. The logarithmic time dependence of isothermal remanent magnetization is observed in ferromagnetic materials with hysteretic magnetization or spin glasses.\textsuperscript{20,22,25,31,32} Generally, it is observed that materials with high coercivity show a pronounced time dependent behavior; however, if the applied field is higher than the coercive field, the relaxation is attributed to spin glass behavior.\textsuperscript{25,31} It may be mentioned that in the case of TbAgAl the relaxation measurements performed after the application of 90 kOe field also exhibit strong relaxation with logarithmic dependence. Therefore, the strong relaxation effects seen in the present case has microscopic (spins) rather than the macroscopic (domains) origin.

Figure 6 shows the zero field temperature dependence of heat capacity ($C$) of TbAgAl. We note that at low tempera-
The temperature dependencies of isothermal magnetization, and heat capacity data of TbAgAl exhibit strong magnetocaloric properties. The magnetization isotherms, temperature and time dependencies of magnetization, and heat capacity data support the notion of glassy state in this compound. The investigations on the magnetocaloric properties reveal that, owing to formation of glassy state, the TbAgAl possesses considerable MCE over a wide temperature span and may find application as a magnetic refrigerant material for refrigeration applications around 60 K.

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FIG. 7. (Color online) Temperature (T) dependencies of isothermal magnetic entropy change (ΔS_M) of TbAgAl calculated for various field changes (ΔH) between 10 and 50 kOe.