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Study of the magnetic phase transitions and magnetocaloric effect in Dy_2Cu_2In compound

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The magnetic properties and magnetocaloric effect (MCE) in Dy_2Cu_2In compound have been investigated. Dy_2Cu_2In undergoes two magnetic phase transitions, a paramagnetic to ferromagnetic (FM) at $T_C \sim 49.5$ K followed by a spin reorientation (SR) at $T_{SR} \sim 19.5$ K. For a magnetic field change of 0–7 T, the maximum values of the magnetic entropy change ($-\Delta S_M^{max}$) are estimated to be 16.5 around T_C and 6.7 J/kg K around T_{SR} with a large relative cooling power (*RCP*) value of 617 J/kg. The modified Arrott plots and universal curves of the rescaled ΔS_M confirmed that the magnetic phase transitions in Dy_2Cu_2In compound belongs the second order phase transitions. The present results may provide some clues to search for new magnetocaloric materials belonging to RE_2T_2X system.

1. Introduction

Magnetic refrigeration based on the magnetocaloric effect (MCE) is an energy-effective and environmental friendly cooling technology compared to the conventional gas compression techniques [1–5]. The MCE is an intrinsic thermodynamic phenomenon for magnetic materials, which can be characterized by the adiabatic temperature change (ΔT_{ad}) or/and isothermal magnetic entropy change (ΔS_M) under a varying magnetic field. Normally, materials with the first order phase transition always possess a large sharp ΔS_M peak around their transition temperatures. However, the first order phase transitions are often accompanied by considerable thermal and/or magnetic hysteresis. In contrast, materials with the second order phase transition always present reversible MCE with a broader temperature range which is beneficial for application. In recent years, much attention has been paid to the rare earth based intermetallic compounds with the second order phase transition, and some of them are found to possess excellent MCE properties that are attractive for active magnetic refrigeration [6–15].

The ternary intermetallic compounds RE_2T_2X (RE = rare earth, T = transition metal, and X = Mg, Cd, Sn or In) crystallized in the tetragonal Mo_2B_2Fe-type structure [16] have attracted some attentions because of their interesting properties, especially for the magnetic behaviours. Depending on the constituent element, the RE_2T_2X compounds undergo various magnetic phase transitions accompanied with a rather wide temperature range of the magnetic transition [17–19]. Among the RE_2T_2X system, only the crystal structure and some basis magnetic characterization in RE_2Cu_2In compounds have been reported [20]. To further understand the physical properties of RE_2Cu_2In compounds, in this paper, the magnetic phase transitions and MCE in Dy_2Cu_2In were investigated in detail.

2. Experimental

High quality polycrystalline sample of Dy_2Cu_2In was prepared by arc-melting appreciate proportions of constituent with the purity better than 99.9% (at.%) under a titanium-gettered argon atmosphere. The sample was melted four times with the button being turned over after each melting to ensure the homogeneity. The

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obtained arc-melted sample was subsequently annealed at 823 K for 7 days in evacuated quartz tubes. The sample was proved to be single phase by X-ray powder diffraction (XRD). The lattice parameters *a* and *c* was evaluated to be 7.466 and 3.746 Å for Dy_2Cu_2In. The magnetic measurements, including temperature and field dependences of magnetization were performed by using a commercial vibrating sample magnetometer (VSM) which is an option of the physical properties measurement system (PPMS-9, Quantum Design) in the temperature range of 3–300 K, with the DC magnetic fields from 0 to 7 T.

3. Results and discussion

Fig. 1 shows the temperature dependence of the zero field cooled (ZFC) and field cooled (FC) magnetization M for Dy_2Cu_2In under various magnetic fields of $H = 0.2$, 0.5 and 1 T. The compound undergoes a paramagnetic to ferromagnetic (PM-FM) transition around the Curie temperature of T_C ~ 49.5 K which is estimated by evaluating the minimum value of the first-order derivative of the magnetization (dM/dT) as a function of temperature. An additional low temperature magnetic transition at T_{SR} ~19.5 K under low magnetic field and shifting to much lower temperature with increasing magnetic field together with some hysteresis is observed which is probably due to the spin reorientation phenomenon often seen on anisotropy crystalline phase [14,21]. These behaviours are consistent with those of previously reported results [20]. Additionally, the ZFC and FC *M-T* curves are well overlapped with each other around and above T_C, indicating no thermal hysteresis as usually observed in magnetic materials with a second order magnetic phase transition which is beneficial for application. Fig. 2 shows the temperature dependence of the magnetization M (left side) and the reciprocal susceptibility $1/\chi$ (right side) for Dy_2Cu_2In under a high magnetic field of 1 T, respectively. The reciprocal susceptibility $1/\chi$ in the PM region obeys the Curie–Weiss law and the paramagnetic Curie temperature θ_p is evaluated to be 48.0 K that confirms the dominant ferromagnetic interactions in the rare earth sublattice for Dy_2Cu_2In compound. The paramagnetic Curie temperature θ_p is deduced from fitting the curve is equal to 10.85 μ_B, which is close to that of free ion value of Dy^{3+} (10.63 μ_B).

The magnetic isothermal $M(H)$ for Dy_2Cu_2In compound is measured in a wide temperature range from 3 to 72 K under applied magnetic field up to 7 T in heating mode, as displayed in Fig. 3(a). It is well known that the magnetic hysteresis during the increasing and decreasing field cycles would lower the cooling power of magnetocaloric materials [22,23]. Therefore, to investigate the magnetic reversibility, the $M(H)$ curves around the T_C and T_{SR} are measured increasing and decreasing field. The magnetization increases sharply under low magnetic fields and shows a tendency to saturate with increasing magnetic field. Moreover, each magnetization isotherm around T_C shows a completely reversible behaviour during the field increasing and decreasing cycles, which is beneficial to practical applications of magnetic refrigeration materials. The Arrott plot curves of the Dy_2Cu_2In compound in the temperature range of 3–72 K are shown in Fig. 3(b). As is well known, according to the Banerjee criterion [24], a magnetic transition is expected to be of the first order if some of the H/M versus M^2 curves show negative slope at some points. On the other hand, it will be of the second order if slopes of all the H/M versus M^2 curves are positive. Only positive slopes can be observed in the Arrott plot curves, indicating magnetic transitions around T_C and T_{SR} are of the second order in nature for Dy_2Cu_2In compound.

Fig. 4 shows the temperature dependence of magnetic entropy change $-\Delta S_M$ for Dy_2Cu_2In compound for different magnetic field

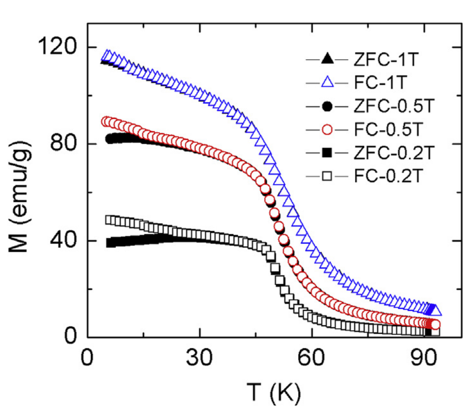

Fig. 1. Temperature dependence zero-field cooling (ZFC) and field cooling (FC) magnetization (M) under the magnetic fields of 0.2, 0.5 and 1 T for Dy_2Cu_2In compound.

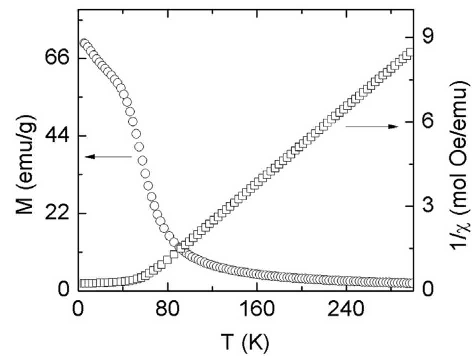

Fig. 2. Temperature dependence of magnetization (M, left scale) and the reciprocal susceptibility ($1/\chi = H/M$, right scale) for Dy_2Cu_2In compound under magnetic field of $H = 1$ T.

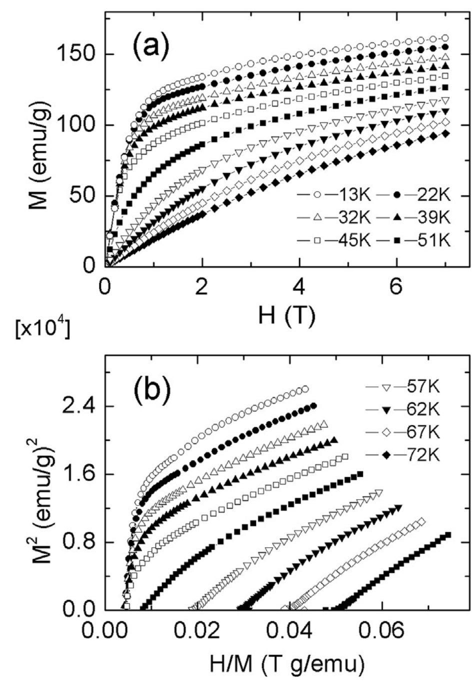

Fig. 3. (a) Magnetic field dependence of the magnetization (increasing field only) for Dy_2Cu_2In at some selected temperatures. (b) The plots of H/M versus M^2 for Dy_2Cu_2In at some selected temperatures.