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Anisotropic magnetocaloric effect in HoAlGa polycrystalline compound

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In this work, a nonnegligible anisotropic magnetocaloric effect (MCE) in HoAlGa polycrystalline compounds has been observed. With temperature increasing, the HoAlGa compound undergoes two kinds of magnetic transitions at 19 K and 31 K, respectively, the later has been recognized as an ordinary antiferromagnetic to paramagnetic (AFM-PM) transition. The -ΔS peak of HoAGa reaches 5.4 J/kg K and 1.5 J/kg K at 35 K along parallel and perpendicular texture directions respectively, for a field change of 0-5 T. The result indicates that the HoAlGa polycrystalline compounds with excellent anisotropic MCE can be expected to have effective magnetic refrigeration applications in low temperature range.
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I. INTRODUCTION

It is well known that magnetic materials impact almost every aspect in our life from household appliances to aerospace field. In recent years, magnetic refrigeration based on the magnetocaloric effect (MCE) has been demonstrated to be a new-style application of functional magnetic materials. Compared with conventional gas compression-expansion refrigeration, magnetic refrigeration has attracted widespread attention due to its energy-efficient and environment-friendly advantages.[1–3] The research and development of magnetic refrigeration utilized at low temperature is very important for the fuel industry and space science.[4] Large anisotropic magnetocaloric effect (MCE) has been acquired in some single crystals, such as $ErGa_2$ and $HoGa_2$, by taking the different entropy change from easy and hard magnetization directions.[5] This idea was first used to study a single-crystal of $DyAlO_3$ by Kuzmin and Tishin.[6] Theoretical and experimental studies performed recently have shown that the use of intrinsic anisotropic properties is capable to improve the magnetocaloric properties of an anisotropic material.[7–10] However, the high cost and complexity of single-crystals growth baffle the widely applications of this kind of magnetic refrigeration model. Zhang Hu et. observed giant rotating MCE in textured DyNiSi polycrystalline material in 2015, for the first time.[11] Rare-earth-based uniaxial intermetallic of RGa_2 compounds have been studied experimentally as well as theoretically.[12–14] Complex field-temperature magnetic phase diagrams of RGa_2 below T_N, characterized by additional transitions, and multistep metal-magnetic processes at low temperature have been reported detailedly.

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Considering the highly preferred crystallographic orientation, a large magnetic anisotropy along different crystalline orientation can be expected in this textured HoAlGa compound. In this work, a nonnegligible anisotropic MCE in HoAlGa polycrystalline compound has been observed due to the texture structure.

II. EXPERIMENTAL DETAILS

Polycrystalline HoAlGa compound was synthesized by arc-melting the stoichiometric mixture of constituent elements Ho, Al, and Ga with high-purity under purified argon atmosphere. 3 at% excessive rare earths were added to compensate the weight loss during the arc-melting. These ingots have been re-melted several times to obtain their homogeneity. Then these samples were annealed in a quartz tube filled with high-purity argon atmosphere for one week at 900 K. Phase purity and crystal structure of annealed samples were checked by Powder X-ray diffraction (XRD) using Cu Ka radiation at room temperature. Magnetizations were carried out on a commercial MPMS SQUID VSM magnetometer (Quantum Design).

III. RESULTS AND DISCUSSION

X-ray diffraction patterns of HoAlGa polycrystalline samples were obtained at room temperature. Almost all the diffraction peaks can be indexed to a hexagonal AlB_2-type structure (Space group P6/mmm), as shown in Fig. 1. The lattice parameters are determined to be a=4.435 Å, b=4.435 Å, and c=3.545 Å. It has been reported that $R(AlxGa_{1-x})_2$ (R=rare earth) compounds crystallize in the simple AlB_2-type hexagonal structure. R atoms lie in the la (0,0,0) site, whereas Al and Ga atoms are randomly distributed on the 2e site.[12–14]

The temperature dependence of magnetization was measured in zero-field-cooling (ZFC) and field -cooling (FC) mode for HoAlGa compounds under the field of 0.01T, as shown in Fig.2. With temperature increasing, the HoAlGa compound undergoes two magnetic transitions at 19 K and 31 K, respectively, which is close to neutron diffraction experiments and Quantitative analysis.[12–14] The peak position at 31K has been recognized as an ordinary antiferromagnetic to paramagnetic (AFM-PM) transition. The ZFC and FC curves are in good consistent with each other, suggesting an excellent thermal reversibility. The inverse susceptibility $1/\chi$ as a function of temperature under 0.01 T is plotted in the inset of Fig. 2. The effective magnetic moment is estimated to be 11.01 μ_B, by simulating the inversed susceptibility vs temperature curve to the Curie-Weiss Law at temperature above 50 K, which is close to the free Ho^{3+}ion value (10.6 μB).

The magnetization isotherms taken below T_N temperature from 0 T to 7 T field are shown in Fig. 3. The significant difference of the slope along parallel and perpendicular texture directions is

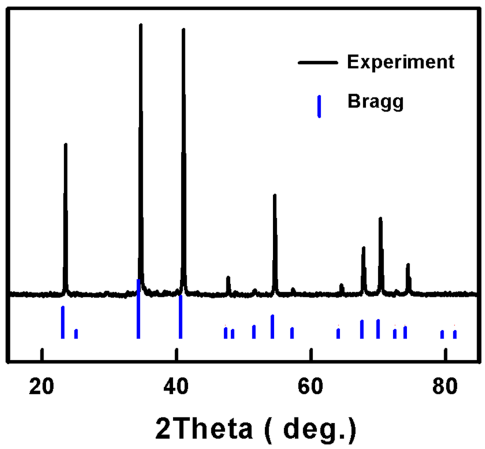

FIG. 1. X-ray diffraction patterns of HoAlGa polycrystalline samples at room temperature. The short vertical lines indicate the angular positions of the Bragg peaks of HoAlGa.

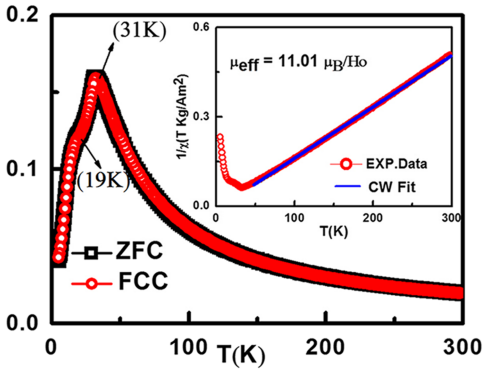

FIG. 2. The temperature dependence of magnetization in zero-field-cooling (ZFC) and field -cooling (FC) mode for TbCoGe compounds under the field of 0.01T. The inset displays the temperature variations of the ZFC inverse susceptibility 1/χ under 0.01 T. The solid line to inverse susceptibility shows the CW fit.

expected to result in a large anisotropy of MCE. Figure 3(b) shows schematic for the formation of texture structure in HoAlGa compound during arc-melting. The formation of texture structure is due to the large temperature gradient between the bottom and top of button during the arc-melting. It was well known that the large rotating MCE in single crystal materials is mainly related to high magneto crystalline anisotropy, as well as large magnetic moments of rare earth ions.[15–17] It is also expected to obtain large anisotropic MCE by rotating the HoAlGa polycrystal sample from perpendicular (90°) to parallel (0°) direction. Along parallel direction, the magnetization curve at 10K shows a linear increase and then a sudden jump with increasing magnetic field, indicating the metamagnetic transition from AFM to FM phase, as shown in figure 3(a). However, there is no magnetic phase change induced by magnetic field along perpendicular direction", suggesting a strong antiferromagnetic coupling.

Magnetic entropy changes of HoAlGa compound in different magnetic fields were calculated using the Maxwell relation $\Delta S_M = \int_0^H \left(\frac{\partial M}{\partial T}\right)_H dH$ based on the magnetization isotherms. Positive value

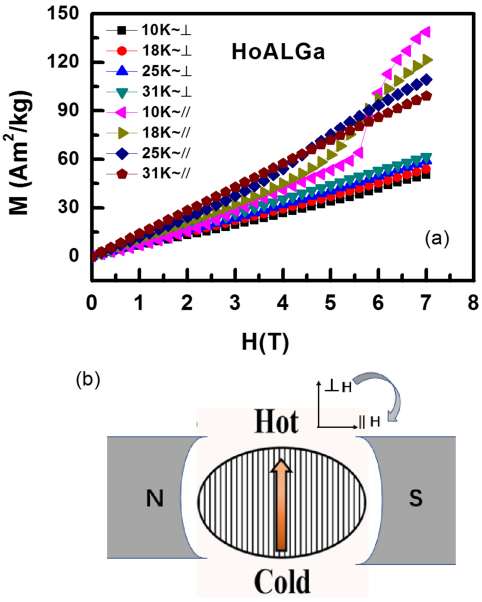

FIG. 3. (a) Magnetization isotherms below T_N along parallel and perpendicular texture directions, respectively. (b) The schematic describes the rotation of sample with texture structure forming during arc-melting from perpendicular (90°) to parallel (0°) direction in magnetic field.

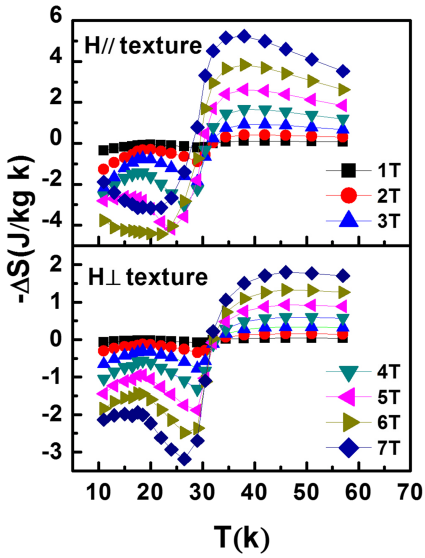

FIG. 4. Magnetic entropy changes of HoAlGa compound along parallel and perpendicular texture directions respectively, under magnetic field changes of 0–1, 2, 3, 4, 5, 6 and 0–7 T.

of ΔS_M were found near 25 K, as shown in Fig. 4, corresponding to the AFM nature of HoAlGa in temperature range from 19 K to 31 K. It is obvious that negative values were also appearing in the latter temperature range, which is reported by neutron diffraction experiments and Quantitative analysis.[13,14] However, we don't observe any abnormality at M-T curves mentioned above. A. R. Ball et. thought it a transition toward the ferromagnetic state. Between 18.5 K and 6 K, the intermediate field-induced phase lead to a two-step metamagnetic process. The maximum values of ΔS_M reach 5.5 J/kg K along parallel texture directions, twice as large as the value of 2 J/kg K along perpendicular texture direction, for the field changes of 0-7 T. The D-value can be compared with the difference between magnetic field applied parallel to the c-axis (a and c) and perpendicular to the c-axis of RGa_2 (R=Ho, Er) single crystal.[5] Above all, a strong magnetic anisotropy for HoAlGa polycrystalline compounds has been observed.

IV. CONCLUSION

AlB2-type hexagonal polycrystalline HoAlGa compound has been synthesized with texture structure in it. With the increasing of magnetic field, HoAlGa compound undergoes two magnetic transitions at 19 K and 31 K, respectively, of which the latter one has been proved to be AFM-PM type magnetic transition. Obvious anisotropy on magnetizing process and magnetic entropy changing was observed due to the strong uniaxial magneto crystalline anisotropy and highly texture structure in HoAlGa compound. The maximum values of $-\Delta S_M$ for 0-7 T were calculated to be 5.5 and 2 J/kgK along texture-parallel direction and texture-perpendicular direction, respectively. Large magnetic anisotropy of textured polycrystalline HoAlGa compound indicates the underlying applications on rotating magnetic refrigeration.

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