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Heat capacity jumps induced by magnetic field in the Er₂HoAl₅O₁₂ garnet



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1. Introduction

Aluminum rare-earth garnets (AlReG), Re₃Al₅O₁₂ (Re – various rare-earth elements), are the most common laser crystalline materials [1,2]. Recently, it was also suggested to use some AlReG in adiabatic demagnetization refrigerators (see [3] and references therein). This stimulated additional interest in caloric properties of AlReG at magnetic field. The heat capacity in AlReG at low temperature is known to be affected by the antiferromagnetic phase transitions and crystal field splitting of the ground energy levels of paramagnetic ions. The latter leads to the Schottky anomalies on the temperature curves of heat capacity (see, for instance, [4,5]). The signature of the emergence of long-range magnetic ordering is a sharp peak in the heat capacity at the phase transition temperature [6] which decreases with increasing magnetic field. In the non-paramagnetic aluminum garnets, $Y_3Al_5O_{12}$ and Lu₃Al₅O₁₂, the heat capacity is totally caused by lattice dynamics [7]. The phonon

ABSTRACT

Measurements of the heat capacity were carried out for the mixed Er₂HoAl₅O₁₂ garnet at magnetic fields up to 15 T. The heat capacity variations at low temperatures were dominated by the Schottky anomalies. In addition, anomalous sharp steps in the heat capacity were observed in magnetic fields stronger than 8 T upon cooling as well as upon warming. The temperatures of the steps increased with increasing magnetic field. Jumps found upon cooling and warming were shifted relative to each other showing the thermal hysteresis. The sharp decrease in the heat capacity at low temperatures suggested the blocking of magnetic filps induced by strong enough magnetic fields.

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contribution to the heat capacity is usually described by combination of the Debye and Einstein terms [8].

Magnetic ordering and behavior in magnetic field of AlReG with paramagnetic ions is influenced by crystalline symmetry. In garnets with the $la\bar{3}d$ symmetry the rare-earth magnetic moments at the dodecahedral c-sites are in the vertices of two interpenetrating triangular sublattices and form a hyperkagome structure [9]. In such a structure the magnetic moments cannot be antiparallel pairwise. This leads to geometric frustration and may potentially give rise to exotic response to the applied magnetic field as, for instance, the magnetic transition in the Tb₃Ga₅O₁₂ garnet [9].

In the present letter we report the anomalous jumps in the heat capacity induced by magnetic field for the mixed aluminum erbium–holmium garnet $\text{Er}_2\text{HoAl}_5\text{O}_{12}$. In this garnet the Er^{3+} and Ho^{3+} magnetic ions occupy the dodecahedral c-sites of the lattice. Until now, no magnetic or heat capacity measurements were carried out for the erbium–holmium mixed garnets as far as we know. The jumps observed show thermal hysteresis upon cooling and warming and move to high temperature with increasing magnetic field. The findings suggest a transformation in magnetic system at strong field which blocks the flips of magnetic moments.

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Fig. 1. Temperature dependences of the heat capacity C at magnetic fields 0, 5, 10, 20, 30, 50, and 90 kOe. The arrow indicates the increasing of magnetic field. The insert shows the temperature dependence of C at zero magnetic field within the enlarged temperature range.

2. Samples and experiment

The $Er_2HoAl_5O_{12}$ single crystal was grown by the horizontal Bridgman directional solidification method in a molybdenum crucible (VNIISIMS, Aleksandrov, Russia). The sample was cut in a shape of plate with faces perpendicular to the crystal cubic axis. The sample mass was 54.19 mg.

The heat capacity was measured with two Physical Property Measurement Systems PPMS-9 + Ever-Cool-II and PPMS-16 from Quantum Design using the built-in conventional procedure. Measurements were carried out within the temperature ranges 300 to 1.9 K in magnetic fields from 0 to 90 kOe and 10 to 2.05 K in magnetic fields from 90 to 150 kOe, respectively. The heat capacity was monitored upon cooling and subsequent warming. In addition, the heat capacity was measured at fixed temperatures of 1.91 and 1.99 K upon gradual increasing magnetic field up to 90 kOe.

3. Results

Temperature dependences of the heat capacity obtained in several magnetic fields from 0 to 90 kOe are shown in Fig. 1 within a reduced temperature range from 30 to 1.9 K. The heat capacity in the whole temperature range from 300 to 1.9 K at zero magnetic field is shown in the inset to Fig. 1. The data in Fig. 1 and the inset were obtained upon warming. The low-temperature behavior of the heat capacity is dominated by the Schottky anomalies. However, one can see an extra step near 2.4 K at the field 90 kOe. Except for a temperature range below this step the heat capacity measured upon warming coincided with that obtained upon cooling. Above about 70 K the heat capacity measured at various magnetic fields almost merge together. The heat capacity curves run close to the curve in the inset and the heat capacity is dominated by the lattice contribution [3,4].

The sharp changes in the heat capacity were found at fields stronger than 80 kOe. The temperature dependences of the heat capacity at large fields observed upon cooling down to the minimal temperature and subsequent warming are shown in Fig. 2. The heat capacity was reversible at the field 80 kOe. At larger fields the sharp steps appeared. The steps emerged upon warming were shifted to high temperature compared to those upon cooling revealing pronounced thermal hysteresis. Measurements at 90 kOe were carried out using both PPMS-9 and PPMS-16 and confirmed the excellent reproducibility of the experimental results. The small



Fig. 2. Temperature dependences of the heat capacity *C* at magnetic fields shown on the panel. The arrows indicate data obtained upon cooling and warming. Thin straight lines show data obtained using PPMS-9 (1) and PPMS-16 (2) at field 90 kOe. The vertical dash line is a guide for the eye as written in the text.



Fig. 3. Temperatures of the steps T_s versus field. Triangles (red online) and circles (blue online) correspond to temperatures obtained upon warming and cooling, respectively. The strait lines are linear fits. Diamonds (purple online) show temperatures of the jumps found at 1.91 and 1.99 K upon increasing field. The dash lines indicate a possible transition temperature at 80 kOe upon cooling.

deviations between positions of the heat capacity jumps seen with PPMS-9 and PPMS-16 arise due to different rates of cooling and warming processes.

The temperatures of the jumps, T_s , which occurred upon cooling and warming increased with increasing magnetic field. The variations of T_s with magnetic field are shown in Fig. 3. Only the data of more accurate measurements with PPMS-9 for the field 90 kOe are included. As can be seen from Fig. 3, the dependences of the jump temperatures on magnetic field are about linear. The linear fits are also shown in Fig. 3.

As the application of magnetic field causes the heat capacity jumps at particular temperatures, one must expect to see similar jumps at a fixed temperature upon changing magnetic field. Fig. 2 shows that the jump at the fixed temperature 1.9 K is expected just between 80 and 85 kOe (see the auxiliary vertical dashed line). Fig. 4 displays the temperature dependence of the heat capacity observed at 1.91 K upon increasing magnetic field from 0 up to 90 kOe with a step of 0.2 kOe. The heat capacity versus field at 1.99 K upon increasing field from 76 up to 90 kOe. The results

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