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Competing magnetic interactions in the intermetallic compound Ho$_2$Mn$_3$Si$_5$


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The compound Ho$_2$Mn$_3$Si$_5$ exhibits multiple magnetic transitions: (i) an ordering at $\sim$78 K, (ii) a second magnetic transition at $\sim$16 K, and (iii) an anomaly at $\sim$4 K. Its paramagnetic Curie temperature is found to be small but positive. Assuming a free ion effective paramagnetic moment of 10.6$\mu_B$ for Ho$^{3+}$ ion, the effective paramagnetic moment per Mn in this compound is calculated to be 1.73$\mu_B$, which indicates the itinerant nature of Mn $d$ electrons. The various transitions in magnetization data are perhaps due to the ordering of rare earth and Mn moments. The magnetization at 2 K in applied fields of up to 7 T has linear field dependence, indicating dominant antiferromagnetic interactions in the system. Neutron diffraction studies point to a complex amplitude modulated incommensurate magnetic structure at 9 K. © 2009 American Institute of Physics. [DOI: 10.1063/1.3072772]

I. INTRODUCTION

Rare earth intermetallic compounds that contain a magnetic lanthanide and a magnetic transition metal are of special interest because they exhibit exotic magnetic ground states, diverse magnetic properties, and complex magnetic structures. In this regard, RMn$_2$ ($R$=rare earth) and RMn$_2$X$_2$ ($X$=Si, Ge) compounds have been extensively investigated to understand the role of $R$-$R$, $R$-Mn, and Mn–Mn interactions on the magnetic properties. The $R_2$Mn$_3$Si$_5$ ($R$=Tb, Dy, Ho, and Er) compounds (tetragonal, space group $P4/mnc$) undergo multiple magnetic transitions because of the magnetic ordering of the two Mn sublattices and one rare earth sublattice. Competing $R$-$R$ and $R$-Mn magnetic interactions in these systems mimic competing interactions in magnetic multilayers. In fact, the $R_2$Mn$_3$Si$_5$ systems exhibit giant magnetoresistance in the magnetically ordered state similar to that often seen in multilayers. In the present work, we examine the intricate magnetic properties of Ho$_2$Mn$_3$Si$_5$ by dc magnetization and low temperature neutron diffraction (ND) measurements.

II. EXPERIMENTAL

A polycrystalline sample of Ho$_2$Mn$_3$Si$_5$ was prepared by arc melting stoichiometric amounts of starting elements (Ho—$99.9\%$ pure; Mn—$99.97\%$ pure; and Si—$99.999\%$ pure) in argon atmosphere. The molten ingot was annealed at 800 °C for 200 h and characterized by room temperature x-ray diffraction. dc magnetization measurements were carried out in the temperature of 1.8–300 K using a commercial superconducting quantum interference device magnetometer (MPMS XL, Quantum Design) in fields of up to 7 T. ND experiments were carried out at 300 and 9 K (incident neutron wavelength 1.4875 Å; MRR, USA). The ND patterns were analyzed using the Rietveld-type computer code FULLPROF.

III. RESULTS AND DISCUSSION

Rietveld analysis of room temperature powder x-ray diffraction data revealed that the Ho$_2$Mn$_3$Si$_5$ is essentially a single phase compound with the tetragonal Sc$_2$Fe$_3$Si$_5$-type crystal structure (space group $P4/mnc$, No. 128). There is a single rare earth site and two crystallographically different Mn sites in each unit cell of this compound. The unit cell lattice parameters are $a=10.595(2)$ Å and $c=5.423(1)$ Å. Trace amounts of AlB$_2$-type HoSi$_{1.66}$ and CeGa$_2$Al$_2$-type HoMn$_2$Si$_2$ were found to be parasitic to the main phase and were included in the refinement.

Temperature dependence of magnetization of Ho$_2$Mn$_3$Si$_5$ in various applied fields is presented in Fig. 1. From the low field magnetization data, it is found that this material orders at $\sim$78 K (denoted as $T_c$ as described below), followed by a second transition at $\sim$16 K (denoted as $T_{K}$). There is a steep increase in magnetization at 78 K as the sample temperature is lowered from the paramagnetic state. This transition at 78 K shifts to slightly higher temperatures on application of larger magnetic fields suggesting the ferromagnetic nature of...
Around this temperature, the magnetization decreases and reaches a minimum at \( T = 16 \) K, after which the magnetization increases again. This indicates the development of competing antiferromagnetic component below 78 K. Application of larger fields enhances the magnetization around \( T_C \) and also leads to the suppression of the competing antiferromagnetic component below 78 K (Fig. 2). However, the transition at 16 K is still discernible even in 1 T field and the magnetization keeps increasing below 16 K, suggesting possible presence of a paramagnetic component. The field dependencies of magnetization data obtained at several temperatures (2–100 K) in fields of up to 7 T reveal nearly linear field dependence (Figs. 3 and 4). A small ferromagnetic component is perceptible at 2, 10, and 40 K in low fields.

The paramagnetic susceptibility of \( \text{Ho}_2\text{Mn}_3\text{Si}_5 \) follows Curie–Weiss law (inset of Fig. 1). The paramagnetic Curie temperature \( (\theta_p) \) is found to be small but positive \( (\sim 0.9 \) K). The positive \( \theta_p \) value again suggests the presence of ferromagnetic interactions in the system. Assuming a full free ion effective paramagnetic moment of \( \text{Ho}^{3+} \) of 10.6\( \mu_B \), the effective paramagnetic moment per Mn is calculated to be 1.73\( \mu_B \) which indicates the itinerant nature of the Mn \( d \) electrons.

The zero-field-cooled and field-cooled magnetization data of \( \text{Ho}_2\text{Mn}_3\text{Si}_5 \), obtained in low applied fields, show considerable thermomagnetic irreversibility below \( T_C \), which is typical of a system with competing magnetic interactions and hence pinning effects. The observation of a subtle but discernible third magnetic transition at temperatures below 4 K in the low field magnetization data of compound may be attributed to the ordering of \( \text{Ho}^{3+} \) moments and requires further detailed study.

Net magnetization value in 7 T field increases from 100 to 10 K and then decreases below 10 K. This observation hints at possible strengthening of the competing antiferromagnetic interactions below 10 K. In general, Mn moments are coupled antiferromagnetically to heavy rare earth moment. This in agreement with the diminishing low field hysteresis as the sample goes from 10 to 2 K (insets of Figs. 3 and 4). The observed magnetic behavior in the title compound could also arise from different temperature dependences of the magnetization of the various magnetic species, namely, \( \text{Ho}^{3+} \), Mn1, and Mn2 moments. Magnetization value reaches a maximum of \( \sim 7.5\mu_B/\text{f.u.} \) at 10 K in 7 T field, which is much less than the value expected from ordered state \( \text{Ho}^{3+} \) and itinerant Mn moments, confirming the dominant antiferromagnetic nature of the material.

Earlier studies on isostructural Tb\(_2\)Mn\(_3\)Si\(_5\) and Er\(_2\)Mn\(_3\)Si\(_5\) compounds also reveal multiple magnetic transitions that evolve with temperature and field in complex manner.\(^{8,9}\) In order to understand the magnetic structure of \( \text{Ho}_2\text{Mn}_3\text{Si}_5 \), ND data were collected at room temperature and at \( \sim 9 \) K (the base temperature of the closed cycle refrigerator used). The ND pattern obtained at 9 K contains a set of incommensurate reflections confirming the dominant antiferromagnetic interactions in this compound at that temperature (Fig. 5). The magnetic structure of this compound appears to be complex, and it could only be understood by performing
more ND experiments as a function of temperature and field and such experiments are planned for the future. Thus the low temperature magnetic structures of $R_2Mn_3Si_5$ ($R=$Tb, Ho, and Er) compounds are somewhat different from each other, suggesting the possible role of single-ion anisotropy of the lanthanide ions in determining overall magnetic properties.

In general, magnetic properties of Mn-containing rare earth intermetallic compounds are ruled by interatomic $R$-Mn and Mn–Mn spacings. The Sc$_2$Fe$_2$Si$_2$-type $R_2Mn_3Si_5$ compounds form from clusters of CeGa$_2$Al$_2$-type, FeSi-type, and AlB$_2$-type compounds and this semieutectic nature could possibly influence their properties. The various bond distances in the title compound are given in Table I. The $R$-$Mn$-2 bonds are metallic, unambiguously ($\Delta$=1), whereas $R$-$R$, $R$-$Mn$1, and Mn1–Mn1 distances are $\sim$1.05–1.10 and hence rare earth sublattice and Mn sublattice orderings could be coupled. Similar behavior has been reported in mixed rare earth containing $RMn_3X_2$ ($X=$Si,Ge) systems where temperature induced ferromagnetic to antiferromagnetic transition in the Mn sublattice is observed giving rise to intermediate magnetic states with varying antiferromagnetic configurations that progressively change their spin orientation as the temperature is changed.$^{10,11}$

**IV. CONCLUSIONS**

The rare earth intermetallic compound Ho$_2$Mn$_3$Si$_5$ shows magnetic transitions at $\sim$78, $\sim$16, and $\sim$4 K. The coupling between rare earth and Mn moments in this system could evolve as a function of temperature. A complex amplitude modulated incommensurate magnetic structure is observed at 9 K.

**ACKNOWLEDGMENT**

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**TABLE I.** Types of atomic bonds in the Sc$_2$Fe$_2$Si$_2$-type $R_2Mn_3Si_5$ compounds: $\Delta=(D_{Atom1-Atom2}^2)/(R_{Atom1}^2+R_{Atom2}^2)$ parameter. Metallic bond if $\Delta$ = 1, covalent or ionic bond if $\Delta$< 1, and van der Waals bond if $\Delta$> 1.

<table>
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<th>$R$</th>
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<th>Mn2</th>
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