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Competing magnetic interactions in the intermetallic compound $\text{Ho}_2\text{Mn}_3\text{Si}_5$ R. Nirmala,¹ A. V. Morozkin,² Jagat Lamsal,³ Z. Chu,⁴ V. Sankaranarayanan,¹ K. Sethupathi,¹ Y. Yamamoto,⁵ H. Hori,⁵ W. B. Yelon,⁶ and S. K. Malik^{7,a)}¹Department of Physics, Indian Institute of Technology Madras, Chennai 600 036, India²Department of Chemistry, Moscow Lomonosov State University, Moscow 119899, Russia³Department of Physics and Astronomy, University of Missouri–Columbia, Columbia, Missouri 65211, USA⁴Department of Physics, University of Missouri–Columbia, Columbia, Missouri 65211, USA⁵School of Materials Science, Japan Advanced Institute of Science and Technology, Ishikawa 923-1292, Japan⁶Department of Chemistry and Materials Research Center, Missouri University of Science and Technology, Rolla, Missouri 65409, USA⁷International Center for Condensed Matter Physics (ICOMP), University of Brasilia, Brasilia 70904-970, Brazil

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The compound $\text{Ho}_2\text{Mn}_3\text{Si}_5$ exhibits multiple magnetic transitions: (i) an ordering at ~ 78 K, (ii) a second magnetic transition at ~ 16 K, and (iii) an anomaly at ~ 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.¹ In this regard, RMn_2 (R =rare earth) and RMn_2X_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.^{1,2} The $\text{R}_2\text{Mn}_3\text{Si}_5$ (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 $\text{R}_2\text{Mn}_3\text{Si}_5$ systems exhibit giant magnetoresistance in the magnetically ordered state³ similar to that often seen in multilayers.⁴ In the present work, we examine the intricate magnetic properties of $\text{Ho}_2\text{Mn}_3\text{Si}_5$ by dc magnetization and low temperature neutron diffraction (ND) measurements.

II. EXPERIMENTAL

A polycrystalline sample of $\text{Ho}_2\text{Mn}_3\text{Si}_5$ 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; MURR, USA). The ND patterns were analyzed using the Rietveld-type computer code FULLPROF.^{5,6}

III. RESULTS AND DISCUSSION

Rietveld analysis of room temperature powder x-ray diffraction data revealed that the $\text{Ho}_2\text{Mn}_3\text{Si}_5$ is essentially a single phase compound with the tetragonal $\text{Sc}_2\text{Fe}_3\text{Si}_5$ -type crystal structure (space group $P4/mnc$, No. 128).⁷ 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 $\text{HoSi}_{1.66}$ and CeGa_2Al_2 -type HoMn_2Si_2 were found to be parasitic to the main phase and were included in the refinement.

Temperature dependence of magnetization of $\text{Ho}_2\text{Mn}_3\text{Si}_5$ in various applied fields is presented in Fig. 1. From the low field magnetization data, it is found that this material orders at ~ 78 K (denoted as T_C as described below), followed by a second transition at ~ 16 K (denoted as T_N). 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

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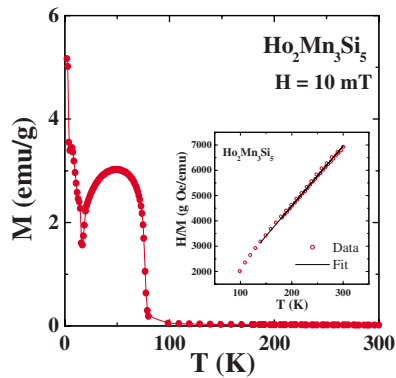


FIG. 1. (Color online) Magnetization vs temperature of $\text{Ho}_2\text{Mn}_3\text{Si}_5$ in 10 mT applied field (inset: M - H isotherm at 40 K).

the transition. A small ferromagnetic component is evidenced from the magnetization versus field isotherm obtained at 40 K. Below T_C , the magnetization decreases and reaches a minimum at ~ 16 K, after which the magnetization increases again. This indicates the development of competing antiferromagnetic component at temperatures of less than 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 (θ_p) is found to be small but positive (~ 0.9 K). This positive θ_p value again suggests the presence of ferromagnetic interactions in the system. Assuming a full free ion effective paramagnetic moment of 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

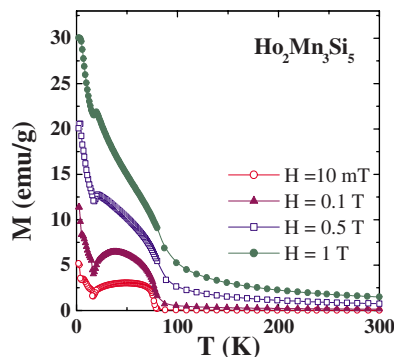


FIG. 2. (Color online) Magnetization vs temperature of $\text{Ho}_2\text{Mn}_3\text{Si}_5$ in various applied fields.

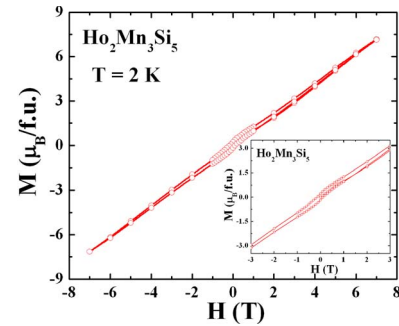


FIG. 3. (Color online) Magnetization vs field isotherm of $\text{Ho}_2\text{Mn}_3\text{Si}_5$ at 2 K in fields of up to 7 T (inset shows the minor loop at 2 K).

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 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 is 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, 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 Ho^{3+} and itinerant Mn moments, confirming the dominant antiferromagnetic nature of the material.

Earlier studies on isostructural $\text{Tb}_2\text{Mn}_3\text{Si}_5$ and $\text{Er}_2\text{Mn}_3\text{Si}_5$ 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 ~ 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

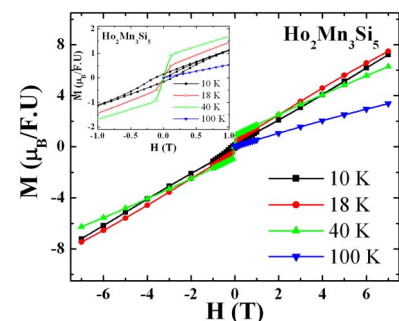


FIG. 4. (Color online) Magnetization vs field isotherms of $\text{Ho}_2\text{Mn}_3\text{Si}_5$ at temperatures of 10, 18, 40, and 100 K (inset: low field magnetization is expanded for clarity).

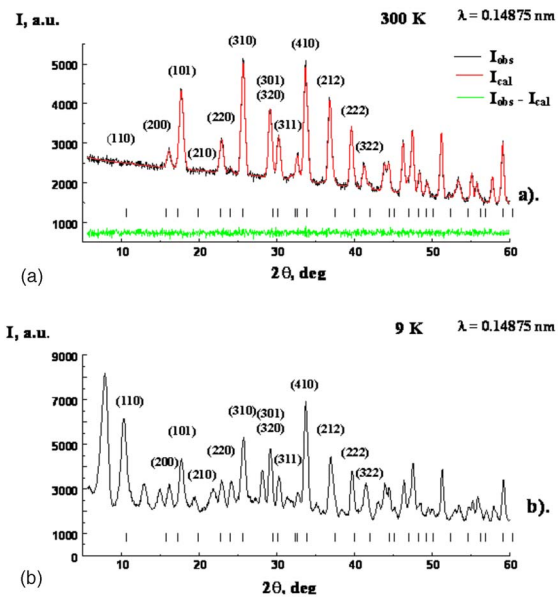


FIG. 5. (Color online) ND patterns of $\text{Ho}_2\text{Mn}_3\text{Si}_5$ compound at 300 (a) and 9 K (b).

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 $R_2\text{Mn}_3\text{Si}_5$ ($R=\text{Tb}$, 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 $\text{Sc}_2\text{Fe}_3\text{Si}_5$ -type $R_2\text{Mn}_3\text{Si}_5$ compounds form from clusters of CeGa_2Al_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 -Mn2 bonds are metallic, unambiguously ($\Delta=1$), whereas R - R , R -Mn1, and Mn1-Mn1 distances are ~ 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_2X_2 ($X=\text{Si}, \text{Ge}$) systems where temperature induced ferromagnetic to antiferromagnetic transition in the Mn sublattice is observed giving rise to in-

TABLE I. Types of atomic bonds in the $\text{Sc}_2\text{Fe}_3\text{Si}_5$ -type $R_2\text{Mn}_3\text{Si}_5$ compounds: $\Delta=(D_{\text{Atom1-Atom2}})/(R_{\text{Atom1}}+R_{\text{Atom2}})$ parameter. Metallic bond if $\Delta \sim 1$, covalent or ionic bond if $\Delta < 1$, and van der Waals bond if $\Delta > 1$.

	R	Mn1	Mn2
R	1.05–1.10	1.04	1.00
Mn1	1.04	1.06	3.28
Mn2	1.00	3.28	1.11

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 $\text{Ho}_2\text{Mn}_3\text{Si}_5$ shows magnetic transitions at ~ 78 , ~ 16 , and ~ 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.

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