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Magnetic phase transitions in $Ho_{11}Pd_4In_9$

Stanisław BARAN[1], Yurii TYVANCHUK[2], Boguslaw PENC[1], Andrzej SZYTUŁA[1]*

[1] *Faculty of Physics, Astronomy and Applied Computer Science, M. Smoluchowski Institute of Physics, Jagiellonian University, Stanisława Łojasiewicza St. 11, PL-30-348 Kraków, Poland*
[2] *Department of Inorganic Chemistry, Ivan Franko National University of Lviv, Kyryla i Mefodiya St. 6, 79005 Lviv, Ukraine*
* *Corresponding author. E-mail: andrzej.szytula@uj.edu.pl*

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The results of ac magnetic susceptibility measurements of $Ho_{11}Pd_4In_9$ are reported. The obtained data show a spin-glass-like anomaly above the critical temperature of long-range magnetic ordering. Two other magnetic phase transitions, related to changes of the long-range magnetic order, are in agreement with previously reported data.

Intermetallics / Magnetic properties / Spin glass / AC magnetic susceptibility

Introduction

Ternary rare earth (R) - transition metal (T) indides with general composition of $R_xT_y\text{In}_z$ exist with different stoichiometries. The compounds have been intensively studied over the past thirty years, due to their complex crystal structures and widely varying magnetic properties [1]. Compounds with the stoichiometry 11:4:9 exist for T equal to Ni [2], Co [3], and Pd [4]. These compounds crystallize in an orthorhombic crystal structure of the $Nd_{11}Pd_4In_9$ type (space group *Cmmm*) [4]. Magnetic measurements indicate that the magnetic moments are localized only on the R^{3+} ions and form complex magnetic structures at low temperatures [5-8]. Particularly interesting magnetic properties have been reported for $Ho_{11}Pd_4In_9$. Its reciprocal dc magnetic susceptibility follows the Curie-Weiss law above 60 K, while noticeable deviations from linearity are observed at lower temperatures (see Fig. 4d in [8]). The ZFC and FC dc magnetic susceptibility curves separate at around 42 K, and below this temperature a fast increase of the FC curve with decreasing temperature is detected. At low temperatures the dc magnetic susceptibility shows a broad peak with the maximum at 15.4 K (ZFC) or 12.3 K (FC).

Neutron diffraction data of $Ho_{11}Pd_4In_9$ disclose the coexistence of two magnetic phases: an antiferromagnetic one described by the propagation vector $\boldsymbol{k}$ = [0, 0, 1/2] and a ferrimagnetic one ($\boldsymbol{k}$ = [0, 0, 0]) at low temperatures. With increasing temperature the antiferromagnetic phase disappears at 11.7 K, while the ferrimagnetic one vanishes around 20 K (see Fig. 8 in [8]).

In order to explain the nature of the magnetic phase transitions in $Ho_{11}Pd_4In_9$, additional ac magnetic measurements have been performed and their results are reported in this work.

Experimental details and results

The measurements were carried out on a sample tested in our previous work [8]. The ac susceptibility was collected using an ACMS option of the Physical Property Measurement System (PPMS) by Quantum Design at a number of frequencies between 100 Hz and 5 kHz at selected temperatures within the temperature range 1.9-60 K. The amplitude of the oscillating magnetic field was equal to H_{ac} = 2 Oe with no direct current field applied (H_{dc} = 0 Oe).

The temperature dependence of the real component of ac magnetic susceptibility (see Fig. 1) shows a broad peak with the maximum at 21 K, and two inflection points at 12 and 30 K. The maximum coincides with the one observed in the dc magnetic susceptibility, while the inflection point at 12 K corresponds to the disappearance of the antiferromagnetic component of the magnetic structure, and the second inflection point around 30 K indicates a transition to the paramagnetic state.

Two maxima, at 16 and 26 K, are found in the temperature dependence of the imaginary component of ac magnetic susceptibility. The temperature of the maximum at 16 K does not depend on frequency and relates to the disappearance of long-range magnetic order. In this temperature region a quick decay of the intensity of the (110)+(020) magnetic peaks is observed (see Fig. 8 in [8]). The temperature of the maximum at 26 K increases with increasing frequency. The origin of this behavior can be understood using the phenomenological classification based on the Mydosh parameter

$$K = \frac{\Delta T_m}{T_m \Delta(\log f)}$$

where T_m is the temperature of the maximum and ΔT_m – the change of this temperature due to a change of the logarithm of frequency equal to $\Delta(\log f)$ [9]. The fit to the experimental data provides the value of K, 0.032, which is comparable to the values reported for typical concentrated spin-glass systems [9,10].

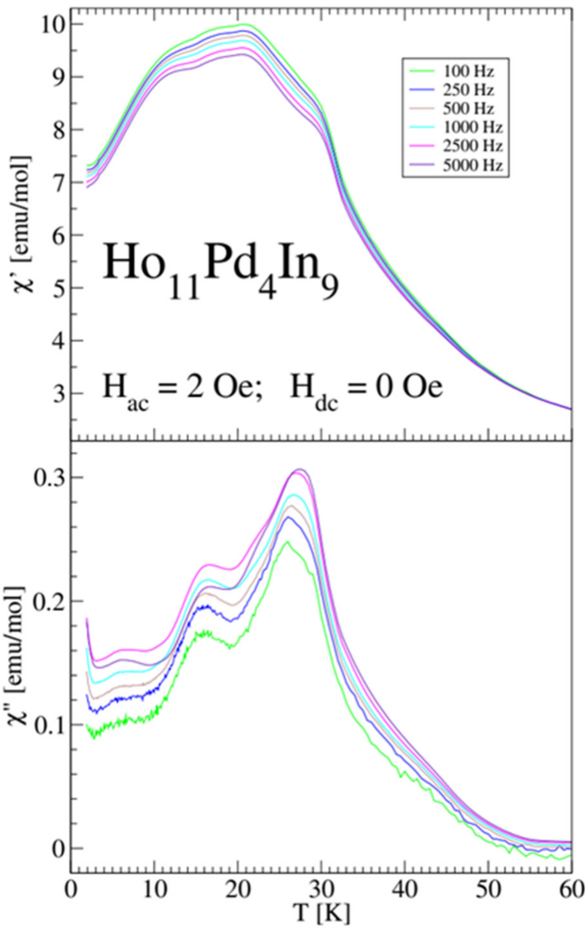

Fig. 1 Ac magnetic susceptibility *vs* temperature for selected frequencies between 100 Hz and 5 kHz. χ' and χ'' refer to the real and imaginary components, respectively.

Discussion

Complex magnetic properties of $Ho_{11}Pd_4In_9$ result from the complex crystal structure of this compound, in which the Ho atoms occupy five non-equivalent sites with different local symmetry, namely *m* for the 2*p* site (Ho1), *m*2*m* for the 4*i* sites (Ho2 and Ho3), 2*mm* for the 4*g* site (Ho4) and *mmm* for the 2*a* site (Ho5). The different Ho atoms have different coordination numbers, namely: Ho1 – 14, Ho2 – 13, Ho3 – 16, Ho4 – 16, and Ho5 – 12.

The investigated compound belongs to a homologous series described by the general formula $R_{m+n}T_{2n}X_m$ with $m = 9$ and $n = 2$. The crystal structure is a stacked one and consists of $n = 2$ fragments of $HoPd_2$ (AlB_2-type) with triangle order of Ho atoms and $m = 9$ fragments of HoIn (CsCl-type) with the atoms forming tetragons (see Fig. 2). The distribution of the Ho atoms in the crystal structure leads to different Ho-Ho interatomic distances: Ho1-Ho1 – 3.454 Å and 3.633 Å, Ho1-Ho2 – 3.488 Å, Ho1-Ho3 – 3.851 Å, Ho1-Ho4 – 3.830 Å, Ho2-Ho5 – 3.481 Å, Ho2-Ho2 – 3.633 Å, Ho3-Ho3 – 3.633 Å, Ho3-Ho4 – 3.978 Å, Ho4-Ho4 – 3.633 Å, and Ho5-Ho5 – 3.633 Å. The values of the shortest Ho-Ho interatomic distances, namely: Ho1-Ho1, Ho1-Ho2, and Ho2-Ho5, indicate that the magnetic interactions in their most have direct character and are described by the Campbell model [11]. The remaining distances are longer than the sum of the atomic radii equal to 3.54 Å, suggesting indirect exchange interactions of the RKKY-type [12]. As a result, a combination of different intra- and inter-sublattices magnetic interactions leads to complex magnetic properties manifesting themselves in the appearance of short-range magnetic order, followed by a transition to long-range order on decreasing the temperature. The spin-glass phase arises from the competition between ferro- and antiferro-interactions [9,10].

Similar properties are observed in TbAuIn. The temperature dependence of the magnetic susceptibility shows two maxima at 33 K and 52 K [13]. Neutron diffraction data indicate long-range magnetic order below 33 K and diffuse neutron scattering in the temperature range 33-52 K is interpreted considering a spin-glass (cluster) system [14].

Summary

The $Ho_{11}Pd_4In_9$ compound with a complex crystal structure, in which the Ho atoms occupy five non-equivalent sites, presents interesting magnetic properties. The results of the ac magnetic susceptibility measurements indicate a change of the magnetic properties with decrease of temperature. A spin-glass-like behavior, observed below 30 K, is followed by a transition to long-range magnetic order at 20 K and a final order-order magnetic transition at 12 K.

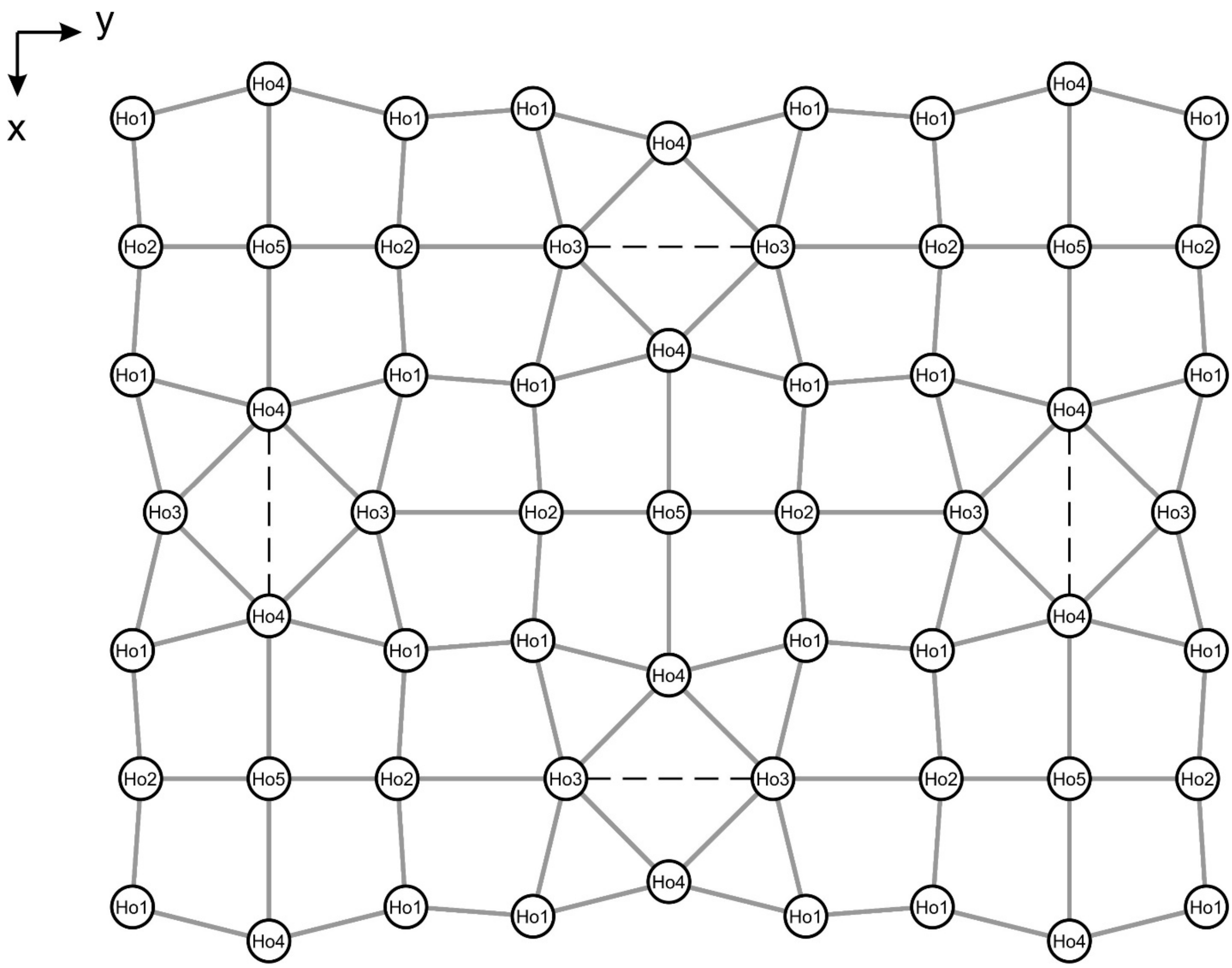

Fig. 2 Distribution of the Ho atoms among the different sites in the structure of $Ho_{11}Pd_4In_9$.

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