Crystal and magnetic structures of $R_2Ni_2In$ compounds ($R = Tb$ and $Ho$)

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Crystal and magnetic structures of $R_2Ni_2In$ ($R = Tb$ and $Ho$) have been studied by powder neutron diffraction at low temperatures. The compounds crystallize in an orthorhombic crystal structure of the $Mn_2AlB_2$-type. At low temperatures, the magnetic moments localized solely on the rare earth atoms form antiferromagnetic structures. The $Tb$ magnetic moments, equal to 8.65(6) $\mu_B$ and parallel to the $c$-axis, form a collinear magnetic structure described by the propagation vector $k = [\frac{1}{2}, \frac{1}{2}, \frac{1}{2}]$. This magnetic structure is stable up to the Néel temperature equal to 40 K. For $Ho_2Ni_2In$ a complex, temperature-dependent magnetic structure is detected. In the temperature range 3.5-8.6 K, an incommensurate magnetic structure, described by the propagation vector $k_1 = [0.76, 0.0, 0.52]$ is observed, while in the temperature interval 2.2-3.1 K the magnetic order is described by two propagation vectors, namely, $k_2 = [\frac{1}{2}, 0.16, \frac{1}{2}]$ and its third harmonics $3k_2 = [\frac{3}{2}, 0.48, \frac{3}{2}]$. Below 2 K, a coexistence of all magnetic structures detected at higher temperatures is observed. For all magnetic phases, the $Ho$ magnetic moments are parallel to the $c$-axis. The low temperature heat capacity data confirm a first order transition near 3 K.

I. INTRODUCTION

There is a strong interest in the development of novel magnetically ordered materials with unconventional properties. The $R_2Ni_2In$ compounds crystallize in the orthorhombic crystal structure (space group $Cmmm$) [1]. Magnetic and specific heat measurements indicate that the $R_2Ni_2In$ ($R = Gd$-$Tm$) compounds are antiferromagnets with the Néel temperatures between 5 K ($R = Tm$) and 40 K ($R = Tb$) [2]. In all compounds, except $R = Ho$, the magnetic order is stable in a broad temperature range between 1.9 K and a corresponding Néel temperature. For $Ho_2Ni_2In$, below the Néel temperature of 9 K, an additional phase transition at $T_1$ equal 3.5 K (dc magnetic data) or 3.3 K (ac magnetic data and heat capacity data) is observed. The more recent paper reports $T_N = 10.5$ K and $T_1 = 5.5$ K [3]. The antiferromagnetic order is confirmed by the neutron diffraction data for $R = Tb$ [3] as well as Er and Tm [3]. The magnetic structure is described by the propagation vectors $k = [1, 1, 1]$ for $R = Tb$ and $[\frac{1}{2}, 0, \frac{1}{2}]$ for Er and Tm. The magnetic moments are found to be localized solely on the rare earth atoms. They form collinear magnetic structure and are parallel to the $c$-axis for Tb and $b$-axis for Er and Tm.

In order to deeper understand magnetic properties of $R_2Ni_2In$ ($R$ - rare earth element), we have performed new neutron diffraction measurements for $R_2Ni_2In$ ($R = Tb$, $Ho$). Although, the magnetic structure of $Tb_2Ni_2In$ has already been reported [3], the previous data have been collected for the sample containing only 10 wt % of $Tb_2Ni_2In$. In the current work we report the results obtained for a new sample consisting of $Tb_2Ni_2In$ in its most part. In addition, we report for the first time the magnetic structure in $Ho_2Ni_2In$. The neutron diffraction data for $Ho_2Ni_2In$ are supported by heat capacity measurements.

II. EXPERIMENTAL DETAILS

The samples of $R_2Ni_2In$ ($R = Tb$ and $Ho$) have been prepared by arc melting of $R$, Ni and In (all with purity at least 99.9 wt %) taken in the atomic ratio of 2:2:1. The melting has been performed under a Ti-gettered Ar atmosphere. The obtained ingots have been turned over and remelted four times in order to get homogeneous distribution of components. The samples have been homogenized in an evacuated quartz-tube at 873 K for one month, followed by cold water quenching.

The samples’ quality has been checked by X-ray powder diffraction at room temperature (PANalytical X’Pert diffractometer with CuKα radiation). Neutron diffraction patterns have been collected in the temperature range from 1.55 up to 60 K on the E6 diffractometer at the Helmholtz-Zentrum Berlin für Materialien in Energie GmbH. The incident neutron wavelength was 2.4315 Å. For Rietveld analysis of the X-ray and neutron diffraction patterns the computer program FullProf has been utilized [4], while for symmetry analysis the computer program Basireps, which is distributed together...

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Heat capacity study have been carried out by a two-tau relaxation method in the temperature range 1.8-12 K using HC option of the Quantum Design PPMS platform.

III. RESULTS

The 2:2:1 compounds crystalize in the orthorhombic crystal structure of the Mn$_2$AlB$_2$-type (space group Cmcm). The atoms occupy the following Wyckoff sites: R at 4j (0, $y_R$, $\frac{1}{2}$), Ni at 4i (0, $y_{Ni}$, 0) and In at 2a (0, 0, 0). The crystal structure of Ho$_2$Ni$_2$In is shown in Figure 3 together with distances between the Ho1 atom and its nearest neighbours. The refined crystal structure parameters at different temperatures are listed in Table I for Tb$_2$Ni$_2$In and Table II for Ho$_2$Ni$_2$In, respectively. The data confirm that the crystal structure is stable down to low temperatures. The shortest interatomic distances between the Ho1 atom and its nearest neighbours in Ho$_2$Ni$_2$In are listed in Table III.

TABLE I. Crystal structure parameters of Tb$_2$Ni$_2$In together with residuals for profile and integrated intensities determined from the neutron diffraction data.

| T [K]     | 60     | 1.55    |
|-----------|--------|---------|
| a [Å]     | 3.913(1) | 3.910(1) |
| b [Å]     | 14.135(2) | 14.125(3) |
| c [Å]     | 3.691(1) | 3.692(1) |
| V [Å$^3$] | 204.15(4) | 203.90(16) |
| $y_Tb$    | 0.3647(2) | 0.3632(3) |
| $y_{Ni}$  | 0.1984(3) | 0.1986(3) |
| $R_{Bragg}$ [%] | 6.2 | 6.6 |
| $R_f$ [%] | 5.0 | 5.8 |

TABLE II. Crystal structure parameters of Ho$_2$Ni$_2$In together with residuals for profile and integrated intensities determined from the neutron diffraction data.

| T [K]     | 20.2 | 3.7 | 2.5 | 1.5 |
|-----------|------|-----|-----|-----|
| a [Å]     | 3.9178(5) | 3.9119(9) | 3.9117(8) | 3.9122(9) |
| b [Å]     | 14.202(2) | 14.184(3) | 14.184(3) | 14.184(3) |
| c [Å]     | 3.6715(4) | 3.6648(8) | 3.6660(7) | 3.6652(8) |
| V [Å$^3$] | 204.29(2) | 203.34(8) | 203.40(7) | 203.38(7) |
| $y_{Ho}$  | 0.3639(4) | 0.3636(6) | 0.3639(6) | 0.3644(6) |
| $y_{Ni}$  | 0.1981(4) | 0.1972(8) | 0.1969(8) | 0.1975(7) |
| $R_{Bragg}$ [%] | 3.4 | 5.1 | 4.9 | 5.3 |
| $R_f$ [%] | 2.8 | 3.6 | 3.0 | 3.2 |

TABLE III. Interatomic distances between the Ho1 atom and its nearest neighbours in Ho$_2$Ni$_2$In determined from the full diffraction patterns.

| distance [Å] | 20.2 K | 3.7 K | 2.5 K | 1.5 K |
|--------------|--------|-------|-------|-------|
| Ho1-Ni2      | 2.825(3) | 2.815(5) | 2.816(5) | 2.821(4) |
| Ho1-Ni1      | 2.986(6) | 2.99(2)  | 3.00(1)  | 2.99(1)  |
| Ho1-In1      | 3.308(3) | 3.306(5) | 3.303(5) | 3.299(5) |
| Ho1-Ho1      | 3.6715(4) | 3.6648(8) | 3.6660(7) | 3.6652(8) |
| Ho1-Ho4      | 3.782(7)  | 3.77(1)  | 3.78(1)  | 3.79(1)  |
| Ho1-Ho2      | 3.866(8)  | 3.87(2)  | 3.86(1)  | 3.85(2)  |
FIG. 2. Neutron diffraction patterns of Ho$_2$Ni$_2$In collected at a) $T = 20.2$ K, b) $T = 3.7$ K, c) $T = 2.5$ K and d) $T = 1.5$ K. Experimental patterns are denoted by points, while the results of Rietveld refinement by lines and the difference curves by bottom lines. From patterns b)-d), the pattern taken at $T = 20.2$ K has been subtracted in order to obtain pure magnetic contribution. The first row of vertical bars indicates Bragg reflection positions originating from the crystal structure of Ho$_2$Ni$_2$In.

The magnetic Bragg reflection positions of Ho$_2$Ni$_2$In related to the propagation vector $k_1 = [0.76, 0, 0.52]$ are denoted by the second row in b) and third row in d). The magnetic Bragg reflection positions of Ho$_2$Ni$_2$In corresponding to the propagation vectors $k_2 = [0.48, 0.16, 0]$ and $3k_2 = [0.48, 0.48, 0]$ are denoted by the second row in c) and d). The nuclear peak positions of HoNi$_2$ impurity are denoted by the second row in a). The magnetic peaks from ferromagnetic order in HoNi$_2$ are indicated by the last rows in b)-d).

Four magnetic $R$ atoms in the $4j$ sublattice with the local symmetry $m2m$ are denoted as: $R_1 (0, yR, \frac{1}{2})$, $R_2 (0, 1 - yR, \frac{1}{2})$, $R_3 (\frac{1}{2}, yR, \frac{1}{2})$, $R_4 (\frac{1}{2}, 1 - yR, -\frac{1}{2})$. The R3 and R4 atom positions are related to the R1 and R2 ones, respectively, by the centering translation $C = [\frac{1}{2}, \frac{1}{2}, 0]$.

The neutron diffraction pattern of Tb$_2$Ni$_2$In, collected at 1.55 K (see Figure 1b), contains additional Bragg reflections originating from the magnetic order. These reflections can be indexed with the use of the propagation vector $k = [\frac{1}{2}, \frac{1}{2}, \frac{1}{2}]$. The same propagation vector describes magnetic order in the isostructural Er$_2$Ni$_2$Pb compound [7]. Symmetry analysis provides four magnetic structure models related to $k$ – for details see Table II in [7]. In case of Tb$_2$Ni$_2$In, the best agreement with the experimental data ($R_{\text{mag}} = 5.0\%$) is obtained for the model T3 with Tb magnetic moments equal to 8.65(6) $\mu_B$ and parallel to the c-axis. The moments follow the $+-+-+$ sign sequence for the Tb1, Tb2, Tb3 and Tb4 atoms in the crystallographic unit cell, respectively.

Temperature dependence of the Tb magnetic moment (see the insert in Figure 1) provides the Néel temperature equal to 40 K, which is in a good agreement with the results of macroscopic magnetic measurements [2].

Neutron diffraction patterns of Ho$_2$Ni$_2$In, collected in the 1.5-20.2 K range, are presented in Figure 2. Ho$_2$Ni$_2$In remains paramagnetic down to 9.1 K. The Bragg reflections of magnetic origin appear in the pattern collected at 8.6 K. With decreasing temperature the Ho$_2$Ni$_2$In magnetic structure undergoes two transitions visible as distinct changes in the diffraction pattern at ca. 3.5 K and ca. 2.0 K, respectively. In order to extract pure magnetic contribution, the paramagnetic pattern taken at $T = 20.2$ K has been subtracted from the patterns recorded at lower temperatures – representative examples are shown in Figures 2b)-d). Propagation vectors describing magnetic structure have been determined using the k-search computer program, while for symmetry analysis of magnetic structures the BasIreps program has been
FIG. 3. Crystal unit cell of Ho$_2$Ni$_2$In (doubled along the $a$-axis for better visibility). The distances between the Ho1 atom and its nearest neighbours are indicated.

utilized. Both programs are parts of the FullProf package [6]. The ferromagnetic contribution arising from the HoNi$_2$ impurity phase has been included in all refinements performed for data collected below 13.5 K [8]. It has been assumed that all magnetic moments in HoNi$_2$ are of the same magnitude and point at the [111] direction.

The Bragg reflections of magnetic origin, observed in the 3.5-8.6 K temperature range, can be indexed with an incommensurate propagation vector $k_1 = [0.76, 0.0, 0.52]$ at $T = 3.7$ K and $k_2 = [5/6, 0.16, 1/2]$ and $3k_2 = [5/2, 0.48, 5/2]$ at $T = 2.5$ K.

The Bragg reflections of magnetic origin, observed in the 3.5-8.6 K temperature range, can be indexed with an incommensurate propagation vector $k_1 = [0.76, 0.0, 0.52]$. Symmetry analysis for $k_1$ and the 4$j$ Wyckoff site provides two irreducible representations: IR1 and IR2. Both representations allow for non-zero contribution to the total magnetic moments along $a$, $b$, $c$ directions (Table IV). All Ho atoms belong to one orbit, therefore they are con-

FIG. 4. Magnetic unit cell of Tb$_2$Ni$_2$In. It is doubled along three directions when compared with the crystallographic one.

FIG. 5. Thermal evolution of neutron diffraction patterns of Ho$_2$Ni$_2$In.

FIG. 6. Modulated magnetic structures of Ho$_2$Ni$_2$In described by the propagation vectors: a) $k_1 = [0.76, 0.0, 0.52]$ at $T = 3.7$ K and b) $k_2 = [5/6, 0.16, 1/2]$ and $3k_2 = [5/2, 0.48, 5/2]$ at $T = 2.5$ K.
TABLE IV. Magnetic structure of Ho$_2$Ni$_2$In described by the IR2 representation for the 4j Wyckoff site of Cmmm space group and propagation vector $k_1 = [k_z, 0, k_z]$. $C_i$ coefficients denote contribution to the total magnetic moment along respective BV$i$ basis vector, $\mu_{tot}$ is the modulation amplitude of magnetic moment and $R_{magn}$ is the magnetic reliability factor of the Rietveld refinement. Only the values for Ho1 and Ho2 atoms are given since Ho3 and Ho4 atoms are interrelated with them by centering translation.

| $k_1 = [k_z, 0, k_z]$ | IR1 BV1 | IR1 BV2 | IR1 BV3 | IR2 BV1 | IR2 BV2 | IR2 BV3 |
|------------------------|---------|---------|---------|---------|---------|---------|
| 4j                     | Ho1     | 0 0 0 0 0 0 1 | 1 0 0 0 | 1 0 0 0 0 0 1 |Ho2     | 0 0 0 0 0 0 1 | 1 0 0 0 | 1 0 0 0 0 0 1 |
| (0, y$\mu_B$, $\frac{1}{2}$) | -1 0 0 0 0 0 0 -1 | 1 0 0 0 | 1 0 0 0 0 0 0 -1 | (0, 1 - y$\mu_B$, $\frac{1}{2}$) | -1 0 0 0 0 0 0 -1 | 1 0 0 0 | 1 0 0 0 0 0 0 -1 |
| $T$                    | Ho$_2$Ni$_2$In | $C_1$ | $C_1$ | $C_1$ | $C_1$ | $C_1$ | $\mu_{tot}$ | $k_z$ | $k_z$ | $R_{magn}$ |
| [K]                    | $[\mu_B]$ | $[\mu_B]$ | $[\mu_B]$ | $[\mu_B]$ | $[\mu_B]$ | $[\mu_B]$ | [\%] |
| 3.7                    | Ho1     | 10.28(9) | 10.28(9) | 0.7580(3) | 0.5198(3) | 5.3 |
|                        | Ho2     | 10.28(9) | 10.28(9) | 0.7580(3) | 0.5198(3) | 5.3 |
| 1.5                    | Ho1     | 8.2(2)   | 8.2(2)   | 0.7568(5) | 0.5191(5) | 5.4 |
|                        | Ho2     | 8.2(2)   | 8.2(2)   | 0.7568(5) | 0.5191(5) | 5.4 |

FIG. 7. Modulation amplitude of the Ho magnetic moments in Ho$_2$Ni$_2$In in function of temperature as obtained by the Rietveld refinement from difference diffraction patterns (representative fits shown in Figures 2b-d). The modulation is described either by the propagation vector $k_1 = [0.76, 0, 0.52]$ or $k_2 = [0.76, 0.16, 0.25]$ and its third harmonics $3k_2 = [\frac{7}{2}, 0.48, \frac{3}{2}]$, indicating that the resultant modulation of magnetic moments differs from purely sinusoidal one, observed at higher temperatures. Symmetry analysis provides identical irreducible representations (IR1 and IR2) for both $k_2$ and $3k_2$. IR1 describes magnetic ordering within the $ab$-plane, while IR2 the one along the c-axis (see Table V). The Ho atoms are divided into two orbits – the first one containing the Ho1, Ho3 pair and the second one containing the Ho2, Ho4 pair. The agreement with experimental pattern is obtained for the IR2 representation (the Ho magnetic moments are parallel to the c-axis). The components of magnetic structure described by the $k_2$ propagation vector and its third harmonics $3k_2$ are visualized in Figure 1b for $T = 2.5$ K. Although the splitting into two orbits allows for different modulation amplitudes and a phase shift between orbits, the Rietveld refinement implies identical modulation amplitudes and zero phase shift between the (Ho1, Ho3) and (Ho2, Ho4) atom pairs.
TABLE V. Magnetic structure of Ho$_2$Ni$_2$In described by the IR2 representation for the 4j Wyckoff site of Cmmm space group and propagation vectors $k_2 = \left[ \frac{1}{2}, 0.16, \frac{1}{2} \right]$ and $3k_2 = \left[ \frac{1}{2}, 0.48, \frac{1}{2} \right]$. $C_1$ coefficient is a contribution to the magnetic moment along BV1 basis vector (the first and second values apply to $k_2$ and $3k_2$, respectively), $\mu_{\text{tot}}$ is the modulation amplitude of magnetic moment, while $R_{\text{mag}}$ is the magnetic reliability factor of Rietveld refinement.

| $T$ [K] | Ho$_2$Ni$_2$In | $C_1$ [$\mu_B$] | $C_2$ [$\mu_B$] | $C_1$ [$\mu_B$] | $\mu_{\text{tot}}$ [$\mu_B$] | $k_y$ | $R_{\text{mag}}$ [%] |
|---------|----------------|-----------------|-----------------|-----------------|-----------------|-------|-----------------|
| 2.5     | Ho1            | 11.41(9), 6.0(2)| 11.41(9), 6.0(2)| 0.1615          | 4.9             |       |                 |
|         | Ho2            | 11.41(9), 6.0(2)| 11.41(9), 6.0(2)|                 |                 |       |                 |
| 1.5     | Ho1            | 6.9(2), 3.5(3)  | 6.9(2), 3.5(3)  | 0.159(2)        | 5.9             |       |                 |
|         | Ho2            | 6.9(2), 3.5(3)  | 6.9(2), 3.5(3)  |                 |                 |       |                 |

The amplitude of modulation related to the $k_2$ propagation vector equals 11.3-11.4 $\mu_B$ in the 2.2-3.1 K range, while the component related to $3k_2$ is about two times smaller and equal to 5.2-6.0 $\mu_B$ (see Figure 6). This relationship between components continues also below 2.2 K, where another magnetic phase transition occurs. It is worth noting that the magnetic structure related to $k_2$ and $3k_2$ resembles the high-temperature magnetic structure related to $k_1$ as both magnetic structures are antiferromagnetic and the magnetic moments are oriented along the c-axis.

At the lowest studied temperatures, a re-appearance of Bragg reflections of magnetic origin, indexed by the $k_1 = [0.76, 0, 0.52]$ propagation vector is observed – notice the presence of magnetic Bragg reflection at $2\theta = 49.3^\circ$ in Figures 2b-d as well as follow thermal evolution of the Ho$_2$Ni$_2$In neutron diffraction pattern shown in Figure 5. A clear co-existence of the magnetic phase described by $k_1$ and that related to the $k_2$, $3k_2$ pair is visible below 2.0 K. The modulation amplitude of magnetic moments related to the $k_1$ propagation vector increases with decreasing temperature reaching finally 8.2(2) $\mu_B$ at $T = 1.5$ K. It exceeds the contributions related to the $k_2$ and $3k_2$ propagation vectors, which decrease at $T = 1.5$ K to 6.9(2) $\mu_B$ and 3.6(4) $\mu_B$, respectively.

In order to get more information about the nature of low-temperature magnetic phase transitions, additional heat capacity measurements have been performed in the temperature range 1.8-12 K with both increasing and decreasing temperature (see Figure 8). A lambda-shaped maximum at 9.0 K is typical of the second-order transition from antiferro- to paramagnetic state, while a distinct thermal hysteresis visible in the temperature range 2.5-3.5 K (see the insert in Figure 5) is characteristic of the first-order transition and coincides with the transition temperature between the magnetic phase described by the $k_1$ propagation vector and that related to the $k_2$ and $3k_2$ vector pair.

FIG. 8. Temperature dependence of the molar heat capacity of Ho$_2$Ni$_2$In in the temperature range 1.8-12.0 K. The insert shows the low-temperature data with increasing (up) and decreasing (down) temperature. The lines are guides for the eye.

IV. DISCUSSION

Analysis of the X-ray diffraction pattern as well as nuclear contribution to the neutron diffraction pattern in function of temperature indicates that the orthorhombic crystal structure is stable in a broad temperature range including the magnetically ordered state (see Tables I and II). The crystal structure is highly anisotropic with the $b$ lattice constant more than three times larger than the $a$ and $c$ ones (see Figure 3). The structure consists of the $ac$-planes of various kinds stacked along the $b$-axis direction, namely, the $R$ atoms planes are intercalated between the Ni- and In-planes following the sequence: In-R-Ni-Ni-R-In-R-Ni-Ni-R-In. The shortest distances between the Ho atoms within the $ac$-plane and those located
at different planes differ (see Table 11) – the lowest are found along the c-axis which is the direction of magnetic moment in the ordered state.

The results reported in this work indicate different kinds of collinear antiferromagnetic orderings present in the investigated compounds, namely, a commensurate one in Tb$_2$Ni$_2$In and an incommensurate one in Ho$_2$Ni$_2$In. In Tb$_2$Ni$_2$In, the low-temperature magnetic structure is described by the propagation vector $k = \left(\frac{1}{2}, 0, \frac{1}{2}\right)$, leading to a magnetic unit cell that has the lattice parameters doubled with respect to the crystallographic cell along all three principal crystallographic directions (see Figure 1). In Ho$_2$Ni$_2$In, a sequence of incommensurate modulated magnetic structures appears with decreasing temperature. Below 9 K a magnetic structure related to $k_1 = [0.76, 0, 0.52]$ is observed, while in the temperature range 2.2 and 3.1 K two propagation vectors ($k_2 = \left[\frac{1}{2}, 0.16, \frac{1}{2}\right]$ and $3k_2$) are required to describe the magnetic structure. Finally, below 2 K a coexistence of both above mentioned magnetic structures is detected.

Temperature-induced transformations of magnetic structures have been found in a number of intermetallic compounds (see Table 1 in [9]). Such transformations of the magnetic structure can be understood on the basis of a realistic mean-field model, which takes into account both periodic-change-field and crystal electric field effects [9]. A sequence of magnetic structures similar to those reported in this work for Ho$_2$Ni$_2$In has been previously observed for UNi$_2$Si$_2$ [10]. With decreasing temperature, an incommensurate modulated magnetic structure ($k = [0, 0, 0.745]$) transforms into an intermediate simple antiferromagnetic one, and finally again to a modulated one ($k = [0, 0, \frac{2}{3}]$) with a ferromagnetic component. Such an evolution of magnetic structure has been interpreted on the basis of the Heisenberg model with biquadratic exchange [11].

The magnetic moments in Tb$_2$Ni$_2$In and Ho$_2$Ni$_2$In are parallel to the c-axis, while they are parallel to the b-axis in R$_2$Ni$_2$In ($R = $ Er and Tm) [9]. Such a change of orientation of the magnetic moment with increasing number of the $4f$ electrons can be attributed to change of sign of the Stevens operator $\alpha_J$ which is negative for Tb and Ho and positive for Er and Tm [12].

In both compound the R-R interatomic distances are about 3.7 Å and therefore they are larger than the sum of respective $R^{3+}$ ionic radii. Such a result suggest presence of indirect exchange interactions of the RKKY-type. The RKKY model predicts proportionality between the Néel temperature and the de Gennes factor defined as $(g_J - 1)^2J(J + 1)$, where $g_J$ is a Landé splitting factor and $J$ is a total angular momentum of the corresponding magnetic ion [13]. Figure 11 in [2] shows a comparison between the experimentally determined Néel temperatures for R$_2$Ni$_2$In ($R = $ Gd–Tm) and those calculated according the RKKY theory. A large discrepancy between the experimental and calculated temperatures for Tb$_2$Ni$_2$In indicates a strong influence of the crystalline electric field (CEF) on magnetic state formation [14]. Therefore the magnetic structures in R$_2$Ni$_2$In ($R$ - rare earth element) result from competition between the RKKY- and CEF-type interactions. Such a competition may lead to complex magnetic properties including temperature-induced magnetic order-order transitions [15]. It is worth noting that such transitions have been observed also in the isostructural R$_2$Ni$_2$Pb ($R = $ Ho, Er) compounds [2, 16]. In Ho$_2$Ni$_2$Pb, a sine-modulated commensurate magnetic order described by the magnetic unit cell $5a \times b \times c$ develops below $T_N = 7$ K. The magnetic structure turns into square-modulated one below $T_i = 3$ K. In Er$_2$Ni$_2$Pb, an incommensurate modulated magnetic structure ($k = [0.8409(1), 0, \frac{1}{2}]$) has been detected below $T_N = 3.5$ K. With decreasing temperature, two intermediate incommensurate magnetic phases, related to the $[0.5973(1), 0, \frac{1}{2}]$ and $[0.5330(3), 0, \frac{1}{2}]$ propagation vectors, respectively, appears. A commensurate magnetic structure, involving two propagation vectors ($[\frac{1}{2}, \frac{1}{2}, \frac{1}{2}]$ and $[0, 0, \frac{1}{2}]$), is found as magnetic ground state.

V. SUMMARY AND CONCLUSIONS

The results presented in this work confirm that the crystal structure of ternary R$_2$Ni$_2$In ($R = $ Tb and Ho) is orthorhombic of the Mn$_2$AlB$_2$-type (space group Cmmm) in both paramagnetic and magnetically ordered states. At low temperatures the rare earth magnetic moments are found to order antiferromagnetically with the rare earth magnetic moments being parallel to the c-axis. In Tb$_2$Ni$_2$In, a commensurate ($k = [\frac{1}{2}, \frac{1}{2}, \frac{1}{2}]$) collinear antiferromagnetic structure with is formed below $T_N = 40$ K. In Ho$_2$Ni$_2$In, a sequence of order-order magnetic transitions is observed. An incommensurate antiferromagnetic sine-modulated structure, related to $k_1 = [0.76, 0, 0.52]$, is detected below $T_N = 9$ K. The structure turns around 3 K into another incommensurate one, which is described by two different propagation vectors, namely, $k_2 = [\frac{1}{2}, 0.16, \frac{1}{2}]$ and $3k_2$. Below 2 K the structure related to $k_1$ reappears and a coexistence of both above mentioned magnetic structures is observed. Heat capacity data reveal that the transition at 3 K is of the first-order type.

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