Coexistence of magnetic fluctuations and long-range orders in the one-dimensional 
$J_1$–$J_2$ zigzag chains materials BaDy$_2$O$_4$ and BaHo$_2$O$_4$

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(Dated: May 15, 2018)

The compounds BaDy$_2$O$_4$ and BaHo$_2$O$_4$ are part of a family of frustrated systems exhibiting interesting properties, including spin liquid-type ground states, magnetic field-induced phases, and the coexistence of short- and long-range magnetic orders, with dominant one-dimensional correlations, which can be described as Ising $J_1$–$J_2$ zigzag chains along the $c$-axis. We have investigated polycrystalline samples of BaDy$_2$O$_4$ and BaHo$_2$O$_4$ with both neutron diffraction and neutron spectroscopy, coupled to detailed crystalline electric field calculations. The latter points to site-dependent anisotropic magnetism in both materials, which is corroborated by the magnetic structures we determined. The two systems show the coexistence of two different long-range orders — two double Néel $↑↑↓↓$ orders in the $ab$-plane with propagation vectors $k_1 = (\frac{1}{2},0,\frac{1}{2})$ and $k_2 = (\frac{1}{2},\frac{1}{2},\frac{1}{2})$ for BaDy$_2$O$_4$, and two distinct arrangements of simple Néel $↑↑↓↓$ orders along the $c$-axis, both with the propagation vector $k_0 = (0,0,0)$ for BaHo$_2$O$_4$. The order for both wave vectors in BaDy$_2$O$_4$ occurs at $T_N = 0.48$ K, while in BaHo$_2$O$_4$, the first order sets in at $T_N \sim 1.3$ K and the second one has a lower ordering temperature of 0.84 K. Both compounds show extensive diffuse scattering which we successfully modeled with a one-dimensional axial next-nearest neighbor Ising (ANNNI) model. In both materials, strong diffusive scattering persists to temperatures well below where the magnetic order is fully saturated. The ANNNI model fits indicate the presence of sites which do not order with moments in the $ab$-plane.

PACS numbers: 75.10.Dg, 75.10.Pq, 75.25.-j, 75.30.Gw, 75.47.Lx, 75.40.-s

I. INTRODUCTION

Among the pyrochlores, spinels, and kagome structures, a newly synthesized family of magnetically frustrated compounds with general formula $AkLn_2O_4$ (where $Ak =$ alkaline earth metal, and $Ln =$ lanthanide) has recently attracted attention 1, as they offer a novel route to magnetic frustration. This family of compounds shows a large variety of magnetic properties, ranging from the coexistence of short- and long-range magnetic order 2–7, to the complete absence of magnetic order down to the lowest accessible temperature despite strong magnetic interactions 8–10. In applied fields, the magnetization shows the formation of plateaus at 1/3 of the saturation value 11, which are concurrent with field induced magnetic order 12–16.

The crystalline structure of the $AkLn_2O_4$ compounds belongs to the $Pnam$ space group, where the lattice of magnetic rare earth ions forms hexagonal tiles in the $ab$-plane, and zigzag chains along the $c$-axis. In this structure, each rare earth ion is located inside a differently distorted O octahedron, resulting in two inequivalent rare earth sites with a distinct crystal electric field (CEF) environment. It has been argued that the unusual magnetism in Sr$Ln_2O_4$ originates from the chains, which can be described as an effective one-dimensional (1D) Ising $J_1$–$J_2$ spin chain model 7, 17 — commonly referred as the 1D axial next-nearest neighbor Ising (ANNNI) model 18. The spin anisotropy in $AkLn_2O_4$ is controlled by the CEF, which splits the $J$ multiplet of the rare earth ions 8. The observed CEF splitting often results in an Ising-type interaction between the rare earth moments, reducing the effective dimension of the exchange to a zigzag chain of spins.

Microscopic models trying to explain ferromagnetic order started out in one dimension, when Ising tried to determine the origin of magnetic order in ferromagnets 19, 20. This development was followed by Heisenberg’s discovery that the origin of the molecular fields in the Weiss theory of ferromagnetism are due to a combination of Coulomb repulsion and the Pauli exclusion principle. At the same time this also gave rise to the possibility of antiferromagnetic interactions, as well as the formation of singlets with spin zero starting from two interacting spin-1/2 21.

The quantum mechanical solution of the 1D antiferromagnetic Heisenberg model was later found by Bethe 22. After this, it took almost 40 years before the remarkable lack of long-range order at finite temperatures for systems with continuous symmetry in one and two dimensions was put on a solid theoretical footing by Mermin
and Wagner.\(^{23}\)

The discovery that the excitations of the 1D spin-1/2 Heisenberg chain are fermions by Faddeev and Takhtajan\(^{24}\), the so-called spinons, led to renewed interest in this field. This work was followed by the seminal papers of Haldane which showed the difference in the excitation spectrum between integer and half-integer 1D chains\(^{25,26}\). The attraction to study low-dimensional spin systems became stronger after the discovery of the high-temperature superconductors which started the search for other such systems in the hope to increase the magnetic fluctuations, and as consequence, the superconducting critical temperature \(T_c\). The result of these efforts, for example, were the so-called ladder compounds by Dagotto and Rice\(^{27}\), which are not only in between one and two dimensions, but also pointed to the important role frustration has in selecting the ground state. Then, Affleck discovered that the application of a magnetic field to integer spin chains in one dimension with antiferromagnetic interactions leads to a quantum phase transition where spin excitations undergo a Bose-Einstein condensation, initiating the search for new quantum magnetic ground states\(^{28}\). Furthermore, the magnetization in these systems shows plateaus, as the magnetization per site is topologically quantized\(^{29}\).

Antiferromagnetic zigzag chains, such as \(AkLn_2O_4\), combine the effect of low dimensionality with magnetic frustration due to the antiferromagnetic next-nearest neighbor interactions. In an applied magnetic fields, zigzag chains show a plateau in the magnetization at 1/3 of the saturation value, even in the classical limit\(^{30}\), and a rich magnetic phase diagram\(^{19}\). Also, while conventional long-range order is prohibited by the Mermin-Wagner theorem, zigzag chains can still acquire chiral order, which should survive to finite temperatures\(^{31}\). In a chirally ordered state, spins have a tendency to rotate in a preferred plane with a preferred rotational direction\(^{31}\).

In the ANNNI model, the ground state at zero temperature is determined through competing interactions between the nearest neighbors \(J_1\) and the next-nearest neighbors \(J_2\):

\[
\mathcal{H} = \sum_i (J_1 \hat{S}_i^z \hat{S}_{i+1}^z + J_2 \hat{S}_i^z \hat{S}_{i+2}^z). \tag{1}
\]

The ANNNI model can accommodate different types of anti-ferromagnetic order, depending on the ratio \(J_2/J_1\) of the interactions\(^{32}\). The ground state is a simple Néel state for an antiferromagnetic \(J_1\) and a \(J_2\) which is either ferromagnetic or weakly anti-ferromagnetic, such that \(J_2 > -0.5|J_1|\). A double Néel state forms for an antiferromagnetic \(J_2\) with \(J_2 < -0.5|J_1|\), where \(J_1\) can be either ferromagnetic, or antiferromagnetic. A ratio close to the critical value separating these two ground states leads to frustration — a strong source for fluctuations.

In this paper, we present a detailed investigation of the magnetic structure of BaDy\(_2\)O\(_4\) and BaHo\(_2\)O\(_4\) for which only growth, specific heat, and magnetization have been reported\(^{33,34}\). The motivation behind this investigation is a direct comparison with the isomorphic compounds Sr(Dy, Ho)\(_2\)O\(_4\) by replacing the non-interacting Sr atoms with larger Ba atoms. As the interactions are governed by crystalline electric field effects, which are very sensitive to local structure, the change from Sr to Ba will change the anisotropy and strength of the magnetic interactions.

II. RESULTS AND ANALYSIS

A. Samples preparation and experimental technique

BaDy\(_2\)O\(_4\) and BaHo\(_2\)O\(_4\) powders were prepared by solid state reaction using high purity starting materials. Stoichiometric mixtures of Dy\(_2\)O\(_3\)/Ho\(_2\)O\(_3\) (99.995\%) and BaCO\(_3\) (99.994\%), with a 1\% surplus of carbonate, both dried at 600°C and weighed in a glove-box, were mixed in a ball mill and pressed into a rod. The rods were heated in an argon atmosphere at 1300°C in an alumina crucible for 12 hours. The solidified rods were ground, repressed into rods, and heated once more under the same condition. This resulted in single phase samples, as determined by X-ray powder diffraction. For specific heat measurements, small single crystals of BaHo\(_2\)O\(_4\) were grown from Ba flux following the method described by Besara et al.\(^{34}\).

In order to establish the CEF splitting of the ground state multiplets \(J = 15/2\) and \(J = 8\) of the Dy\(^{3+}\) and the Ho\(^{3+}\) ions, respectively, inelastic neutron scattering experiments were performed on powders of both BaDy\(_2\)O\(_4\) and BaHo\(_2\)O\(_4\) using the MERLIN time-of-flight spectrometer at ISIS in the United-Kingdom\(^{35}\). The two samples were mounted in a double-walled aluminum cylinder to maximize the scattering intensities\(^{36}\). Spectra were acquired for both samples at incident neutron energies \(E_i\) of 12.30, 20.00, 38.20 and 100.00 meV and temperatures \(T\) of 7, 75 and 150 K. A calculation of the CEF energy levels and the inelastic neutron spectra were performed with the program MULTIX\(^{37}\).

To determine the magnetic structure of the compounds, elastic neutron scattering data were acquired on the same powders using the HRPT diffractometer at the Paul Scherrer Institut (PSI) in Switzerland\(^{38}\). Data were collected from \(T = 0.07\) K up to 30 K at a neutron wavelength \(\lambda = 1.886\ \text{Å}\) for BaDy\(_2\)O\(_4\), and from \(T = 0.20\) K up to 60 K at the same wavelength, with additional data at \(\lambda = 1.155\ \text{Å}\) for BaHo\(_2\)O\(_4\). Due to the strong neutron absorption of Dy, the BaDy\(_2\)O\(_4\) powder was loaded into a double walled copper cylinder, while for BaHo\(_2\)O\(_4\), a simple copper cylinder was used. Both cylinders were filled with 10 bar of helium at ambient temperature to ensure a good thermalization. The magnetic structure was refined through a Rietveld analysis with the FULLPROF Suite\(^{39}\).

Magnetization measurements on the powders were carried out using a Quantum Design SQUID-VSM, equipped with a 7 T magnet, over the temperature range from 300 K down to 1.8 K. The specific heat measurements...
Fig. 1. Inelastic neutron scattering spectra for BaDy$_2$O$_4$ (top) and BaHo$_2$O$_4$ (bottom) at $T = 7$ K for various incident energies $E_i$. Here, the spectra have been summed over all $|Q|$ values. The solid line is the sum of the contributions from the two rare earth sites. The contribution from site 1 is shown as the orange dashed line and the contribution from site 2 is shown as the blue dashed line. The lines were obtained by fitting the experimental spectra with calculations using the program MULTI$\times$.

were made using a Quantum Design PPMS equipped with an $^3$He refrigerator, down to a temperature of 0.35 K.

**B. Crystalline electric fields excitations**

The program MULTI$\times$ was used to calculate the inelastic neutron spectra for comparison with the experimental spectra. It computes the energy levels of an individual magnetic ion in the CEF defined by the charges at the positions of the neighboring ions. This permits the characterization of the single-ion anisotropy. Here, we are taking advantage of the fact that MULTI$\times$ allows us to calculate CEF levels in systems with low symmetry, such as the BaLn$_2$O$_4$ structure, where the only point symmetry at the rare earth site is a mirror plane.

Beside the CEF, the energy levels are calculated by MULTI$\times$ by taking into account the Coulomb and spin-orbit interaction. The strength of these three effects can be scaled in the model through semi-empirical parameters. The computed spectra are fairly insensitive to the scaling of the strength of the Coulomb and spin-orbit interaction of the Hamiltonian. In consequence, only the CEF scaling factor $S_{\text{CEF}}$ was refined, resulting in a fair agreement with the experimental spectra, as shown in Fig. 1. For these spectra the data for all wave vectors $|Q|$ was summed. The calculations were carried out independently for the two crystallographically inequivalent rare earth sites, with a different CEF scaling factor for each site. In order to compare the calculations with the inelastic neutron spectra, the contribution from both sites was added. For BaDy$_2$O$_4$ the best fit was obtained with $S_{\text{CEF}} = 0.19$ and $S_{\text{CEF}} = 0.64$, and for BaHo$_2$O$_4$ with $S_{\text{CEF}} = 0.45$ and $S_{\text{CEF}} = 0.58$. These low values of the $S_{\text{CEF}}$ parameters are similar to what has been observed in Sr(Dy, Ho)$_2$O$_4$,

| $\mu_a$ | $\mu_b$ | $\mu_c$ | $|\mu|$ |
|--------|--------|--------|-------|
| Dy$_{1\text{CEF}}$ | 9.4 | 2.4 | 0.1 | 9.7 | Dy$_{1\text{exp}}$ | 2.9 | 4.2 | 0.2 | 5.1 |
| Dy$_{2\text{CEF}}$ | 1.9 | 9.6 | 0.0 | 9.8 | Dy$_{2\text{exp}}$ | 0.0 | 2.3 | 0.0 | 2.3 |
| Ho$_{1\text{CEF}}$ | 1.2 | 9.6 | 0.0 | 9.7 | Ho$_{1\text{exp}}$ | Not ordered |
| Ho$_{2\text{CEF}}$ | 0.0 | 0.0 | 7.9 | 7.9 | Ho$_{2\text{exp}}$ | 0.0 | 0.0 | 6.2 | 6.2 |

and from a refinement of the magnetic Bragg peaks $E_i = 12$ meV for the Dy$^{3+}$ site, respectively, while for BaHo$_2$O$_4$ these levels are much closer to the ground states, with $E = 0.2$ meV and 0.3 meV for the Ho$_1^{3+}$ and Ho$_2^{3+}$, respectively. A comparison between the energy levels obtained by fitting the inelastic neutron scattering spectra and the position of the peaks in the spectra is shown in Fig. 2. Here, the positions of the experimental levels were determined by fitting Gaussian functions to the peaks in the spectra.

The MULTI$\times$ calculations also predict the size and direction of the magnetic moments. Each Dy$_{3+}$ ion has a doublet ground state protected by time reversal symmetry due to the fact that Dy$_{3+}$ is a Kramers ion. For each Ho$_{3+}$, which is a non-Kramers ion, the ground state is treated as a pseudo-doublet because the lowest energy states are two singlets separated by an energy difference smaller than the computational accuracy. The moments obtained by this procedure are listed in Table I. For BaDy$_3$O$_4$, the moments for both sites lie in the $ab$-plane, with one predominantly along the $a$-axis and the other along the $b$-axis. For BaHo$_2$O$_4$, the first site has a moment in the $ab$-plane, while for the second site, the moment is along the $c$-axis. There is a clear distinction between moments laying exclusively in the $ab$-plane or exclusively along the $c$-axis — perpendicular or along the chains. This points to a clear anisotropy of the magnetic interactions, as previously observed for the Sr variant of these compounds. We observe the following total moments per each site: $|\mu_{\text{Dy}_1}| = 9.7\mu_B$, $|\mu_{\text{Dy}_2}| = 9.8\mu_B$, $|\mu_{\text{Ho}_1}| = 9.7\mu_B$, and $|\mu_{\text{Ho}_2}| = 7.9\mu_B$. Of these, only
the second Ho site has a moment which is significantly lower than the effective moment expected from Hund’s rules — 10.4µB for both Dy^{3+} and Ho^{3+} ions. Fitting a Curie-Weiss model to the magnetic susceptibilities at high temperatures results in an effective moment µ_{eff} per magnetic site of 10.60(1)µB and 10.77(1)µB for BaDy_2O_4 and BaHo_2O_4, respectively, with Curie-Weiss temperatures θ_{CW} of −18.5(3) K and −10.9(3) K.

The calculations of the CEF levels with MULTIX are fairly sensitive to the details of the crystallographic structure. The low symmetry of the structure is the source of this complex level scheme, and slight distortions of the structure could have a great impact on the results, as this calculation is highly dependent on the atomic positions. It is then important to determine whether any significant distortion occurs upon cooling. For this, we refined the lattice constants and the atomic positions within the unit cell from the neutron diffraction spectra at high angles 2θ, as the magnetic scattering intensity is stronger at low angles. For BaHo_2O_4, a simultaneous refinement of the data acquired at wavelengths λ of 1.886 Å and 1.155 Å was performed. Only a small variations of the O distances is observed in the entire range of temperatures from 60 K down to 0.07 K, as shown in Fig. 3. The chemical unit cell volume varies by about 0.5 Å³ for BaDy_2O_4, and there is no volume change for BaHo_2O_4 within the experimental accuracy. The slight change in the lattice parameters of BaDy_2O_4 is accompanied by a small increase of the Bragg peaks’ width. The intra-chain distance between the rare earth atoms is also temperature independent and not affected by the magnetic order. The dimension of the O octahedra surrounding the magnetic ions have the biggest influence on the CEF splitting, and a thermal distortion would affect the single-ion anisotropy. However, no such distortion is observed for both BaDy_2O_4 and BaHo_2O_4. Such a distortion was previously noted in the structurally equivalent compounds of SrTb_2O_4 and SrTm_2O_4, but the change in these compounds is rather subtle and below our experimental resolution. For BaDy_2O_4, the lattice parameters are a = 10.4068(3) Å, b = 12.1131(3) Å and c = 3.46985(10) Å at T = 0.07 K, and for BaHo_2O_4, a = 10.3864(3) Å, b = 12.0852(2) Å and c = 3.44754(8) Å at T = 0.20 K. These values are similar to those reported by Doi et al.

The CEF level scheme of BaDy_2O_4 was robust to repositioning the atoms within the error bars of our structural refinement and only a scaling of the level separation was observed. However, in BaHo_2O_4, the level scheme was more strongly affected, and significantly changes when the O positions are set to the limit of the uncertainties of our refinement. Despite the absence of clear structural changes, the MULTIX calculations would nonetheless benefit from higher resolution data from a X-ray synchrotron source to observe potential distortion of the O octahedra.
netic order with two propagation vectors \( \mathbf{k} \) of BaDy. The top panel shows the magnetic peaks at low diffraction angles. The top panel shows the magnetic peaks at \( \theta \) of BaDy's of 16°, 17°, and 18°.

Additional measurements are planned, such as electron paramagnetic resonance (EPR) and photoluminescence spectroscopy, as were used in the case of SrEr₂O₄ to determine the CEF levels.

The motivation for carrying out these CEF calculations was to establish a basis for the characterization of the magnetic structures in the following sections. The results from the CEF model for the magnetic anisotropy and direction of the easy-axis are important for developing a magnetic model.

C. Magnetic structure

1. BaDy₂O₄

Information related to the ordering temperature as well as the nature of the magnetic order can be deduced from the temperature dependence of the powder diffraction spectra. Spectra at low diffraction angles \( \theta \) from base temperature to \( T = 1 \) K are shown in Fig. 4. In the case of BaDy₂O₄, upon cooling below \( T = 0.48 \) K, additional Bragg peaks appear at \( \theta \)'s of 16.5°, 17.1°, and 18.8°. These peaks can be indexed with a momentum transfer \( \mathbf{Q} \) of \( (\frac{1}{2},0,\frac{1}{2}), (\frac{1}{2},\frac{1}{2},\frac{1}{2}) \), and \( (\frac{1}{2},1,\frac{1}{2}) \), which indicate a magnetic order with two propagation vectors \( \mathbf{k}_1 = (\frac{1}{2},0,\frac{1}{2}) \) and \( \mathbf{k}_2 = (\frac{1}{2},\frac{1}{2},\frac{1}{2}) \).

On a wider range of angles, as shown in Fig. 5(a)–(b), one can notice the evolution of an additional magnetic scattering signal when the magnetic transition temperature is approached. This additional signal has the form of a sawtooth, which is reminiscent of the powder average for one-, or two-dimensional magnetic correlations. This diffuse scattering is present in both compounds, and contrary to what one would expect for a fully ordered system, it does not completely disappear with the onset of the magnetic Bragg peaks, but remains visible down to the lowest measured temperatures. The main feature of
of the magnetic Bragg peaks indicates that the order is different ordering vectors are each associated with crystallographically different rare earth site. The sharpness of the magnetic Bragg peaks indicates that the order is long-range, as their widths are comparable to the nuclear Bragg peaks. A visualization of the structures is given in Fig. 7.

Each of the two magnetic sites forms a chain which has a double Néel structure with a ↑↑↓↓ motif. As noted in Fig. 4, site 1 has a propagation vector \( \mathbf{k}_1 = (\frac{1}{2},0,\frac{1}{2}) \) and site 2 has \( \mathbf{k}_2 = (\frac{1}{2},\frac{1}{2},\frac{1}{2}) \), quadrupling and octupling the volume of the chemical unit cell. Both ordering vectors correspond to the same Shubnikov group with different irreducible representations (irreps) of the space group \( Pnam \), as listed in Table II. Chain A has a ↑↑↓↓ motif for both sites. In contrast, on chain B, one site has a ↑↑↓↓ motif, the other a ↓↑↑↑↓ motif.

The structure was refined for all measured temperatures, which made it possible to extract the temperature dependence of the size and direction of the magnetic moments. For this analysis, the scaling parameters in the refinement were fixed to the one from the nuclear Bragg peaks intensity, in order to obtain the correct moment size. The results for both propagation vectors and irreps are shown in Fig. 8.

From the refinement, we know that both moments are lying in the \( ab \)-plane: \( \mathbf{\mu}_{k_1} = (2.9,4.2,0.0)\mu_B \) and \( \mathbf{\mu}_{k_2} = (0.0,2.3,0.0)\mu_B \). Also, both moments order simultaneously at \( T_N = 0.48 \) K, within the temperature resolution of our experiment, and they saturate already by 0.30 K.

From the analysis of the powder spectra, it is not possible to assign the different wave vectors to a particular rare earth site — the \( \chi^2 \) from the refinements are not significantly different when we exchange the sites, or if both wave vectors are confined to a single site. Nonetheless, identification of the sites is possible by comparing the direction of the magnetic moments obtained from the CEF calculations for the different crystallographic sites, listed in Table I, with the direction of the moments from the refinement of the powder spectra. For \( \text{BaDy}_2\text{O}_4 \), the MULTIX calculations and the refinement from the experimental diffraction pattern both show that both moments are restricted to the \( ab \)-plane. We can now use the relative strength of the components along \( a \) and \( b \) to distinguish between the two sites — the moment \( \mathbf{\mu}_{\text{Dy}_1} \) is stronger along \( a \) and the moment \( \mathbf{\mu}_{\text{Dy}_2} \) stronger along \( b \). The resulting magnetic structure is shown in Fig. 7, where we show the different sites and chains separately for clarity. The discrepancies between the size of the magnetic moments from the refinements and those predicted by the CEF calculations can be explained by the presence of diffuse scattering, analyzed in Sec. II.D.

2. \( \text{BaHo}_2\text{O}_4 \)

For \( \text{BaHo}_2\text{O}_4 \), new peaks are observed for temperatures below \( T = 0.84 \) K at values of \( Q \) corresponding to a propagation vector \( \mathbf{k}_0 = (0,0,0) \). They are clearly visible at low angles in Fig. 4. Additional intensity also appears at the position of nuclear Bragg peaks at higher
TABLE II. Basis vectors of the irreducible representations of the space group $Pnam$ (No. 62), where the magnetic ions occupy position 4c which has a $m$ symmetry, for each propagation vector $\mathbf{k}$ of the magnetic structures of BaDy$_2$O$_4$ and BaHo$_2$O$_4$.

| BaDy$_2$O$_4$ | $\mathbf{k}_1 = (\frac{1}{2}, 0, \frac{1}{2})$ | $\mathbf{k}_2 = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ |
|---------------|---------------------------------|---------------------------------|
| $x,y,z$       | $(C_1 - iC_4, C_2 - iC_5, C_3 + iC_6)$ | $(C_1 - iC_4, C_2 - iC_5, C_3 + iC_6)$ |
| $-x,-y,z+\frac{1}{2}$ | $(-C_4 + iC_1, -C_5 + iC_2, -C_6 - iC_3)$ | $(-C_4 + iC_1, -C_5 + iC_2, -C_6 - iC_3)$ |
| $x+\frac{1}{2},y+\frac{1}{2},z+\frac{1}{2}$ | $(-C_4 - iC_1, C_5 + iC_2, -C_6 + iC_3)$ | $(C_4 + iC_1, -C_5 - iC_2, -C_6 + iC_3)$ |
| $-x+\frac{1}{2},y+\frac{1}{2},z+\frac{1}{2}$ | $(C_4 + iC_1, -C_5 - iC_2, -C_6 + iC_3)$ | $(C_4 + iC_1, -C_5 - iC_2, -C_6 + iC_3)$ |
| BaHo$_2$O$_4$ | $\mathbf{k}_{11} = (0,0,0)$ | $\mathbf{k}_{12} = (0,0,0)$ |
| $x,y,z$       | $(0,0,C_1)$ | $(0,0,C_1)$ |
| $-x,-y,z+\frac{1}{2}$ | $(0,0,-C_1)$ | $(0,0,-C_1)$ |
| $x+\frac{1}{2},y+\frac{1}{2},z+\frac{1}{2}$ | $(0,0,C_1)$ | $(0,0,-C_1)$ |
| $-x+\frac{1}{2},y+\frac{1}{2},z+\frac{1}{2}$ | $(0,0,-C_1)$ | $(0,0,-C_1)$ |

diffraction angles.

The diffuse scattering in Fig. 5(c)–(d) is similar to the case of BaDy$_2$O$_4$. However, it is easier to see in BaHo$_2$O$_4$ with its $\mathbf{k}_0$ propagation vector, where once again the diffuse signal is stronger at $29 \sim 18^\circ$. This diffuse scattering signal is still visible on the otherwise stable background up to $T = 30$ K, but also persist below the magnetic transition down to 0.20 K where the feature becomes much sharper.

The result of the refinement of the diffraction patterns is shown in the lower panels of Fig. 6. Like its sister compound BaDy$_2$O$_4$, BaHo$_2$O$_4$ possesses two distinct magnetic orders, characterized by sharp resolution limited magnetic Bragg peaks. In this case, they consist of simple Néel chains $\uparrow\downarrow\uparrow\downarrow$ with a propagation vector $\mathbf{k}_0 = (0,0,0)$, which does not increase the size of the unit cell. Two magnetic orders with different irreps, $\Gamma_1$ and $\Gamma_2$, are nonetheless needed in order to adequately account for magnetic scattering intensities. With such propagation vector, each possible irrep is forced by symmetry to either carry a moment in the $ab$-plane or along the $c$-axis, whereas the irreps of $\mathbf{k}_1$ and $\mathbf{k}_2$ of BaDy$_2$O$_4$ are not restraining the moment direction.

Moreover, a close inspection of the data together with the refinement, presented in the lower panels of Fig. 6, show that the two orders — or sets of Bragg peaks — appear in two steps. The contribution from $\Gamma_1$ indicates order at 0.84 K, and the one from $\Gamma_2$ orders between 1 K
and 2 K, a temperature range in which unfortunately no additional was gathered. Both orders share the same propagation vector and have ordered moments perpendicular to the \(ab\)-plane, pointing along \(c\). Interestingly, this points to a coexistence of the ordered moments on site 2, leaving the moment on site 1 disordered. This is then consistent with the \textsc{multiX} calculations which show that the moment of the Ho on site 2, \(\mu_{\text{Ho}_2}\), points along \(c\) while the moment on site 1, \(\mu_{\text{Ho}_1}\), is restricted to the \(ab\)-plane. There are several possibilities to interpret this structure from the powder diffraction refinement. One rather unusual interpretation is to treat the two orders as distinct phases. This would imply the superposition of the two orders on the same site, resulting in magnetic moments on chains A and B with starkly different sizes.

The more physical scenario is that the correlations in the chains nucleate either one of the two magnetic structures, leading to a phase separation within a single chain. In this case, the same moment is placed on both irreps, while the phase fraction is allowed to vary. The refinement of \(\text{BaHo}_2\text{O}_4\) yields a total moment \(\mu_{k_0} = (0.0, 0.0, 6.2)\mu_B\), where the moment increases with decreasing temperature in two steps, as seen in the lower panel of Fig. 8. The inset shows the phase fraction of each irrep, normalized to 1. From \(T_{\text{NF}_2} \sim 1.3\) K down to the second ordering temperature \(T_{\text{NF}_1} = 0.84\) K, only \(\Gamma_2\) contributes to the magnetic diffraction. At this temperature, a new set of Bragg peaks characterized by \(\Gamma_1\) appears. For temperatures below the onset of the new Bragg peaks, near \(T = 0.75\) K, the phase fraction stabilizes at a ratio 80% of \(\Gamma_1\) and 20% of \(\Gamma_2\). This conclusion is also supported by our analysis of the diffuse scattering intensity, detailed in the next section.

Previous specific heat measurements on powder of \(\text{BaHo}_2\text{O}_4\) report only one antiferromagnetic phase transition at \(T_N = 0.8\) K\textsuperscript{33}. This is in contradiction with our own measurements which show a transition at \(T_N = 1.2\) K, which is close to \(T_{\text{NF}_2} \sim 1.3\) K, associated with the linear extrapolation of the moment size in Fig. 8. The specific heat measurements shown in Fig. 9 were performed on small single crystals obtained through flux growth\textsuperscript{34}. The large increase seen at the lowest temperatures is the signature of a nuclear Schottky contribution \(C_{\text{nc}}\). Subtracting \(C_{\text{nc}}\) from the measured data yields the magnetic contribution \(C_{\text{mag}}\), as in this temperature range, the lattice contribution is negligible. The entropy \(S_{\text{mag}}\) is calculated by integrating \(C_{\text{mag}}(T)/T\). While the temperature range is too narrow to show a saturation, it is interesting at this point to note that the magnetic transition has only a very small contribution to the entropy of the system. Just above \(T_N = 1.2\) K, it is well below the expected value of \(R \ln(2)\) for the ordering of a doublet, where \(R\) is the universal gas constant. This indicates very little difference in entropy between the fluctuating chains and long-range magnetic order.

![FIG. 8. Size and direction of the magnetic moment as determined from the refinement of the neutron powder diffraction data. In \(\text{BaDy}_2\text{O}_4\), the moments for both wave vectors \(k_1\) and \(k_2\) order simultaneously at \(T_N = 0.48\) K. In \(\text{BaHo}_2\text{O}_4\), the two irreps, which have the same wave vector \(k_0 = (0.0, 0.0)\), order at different temperatures. The dotted lines are linear extrapolations. The inset shows the phase fraction of the two irreps, where \(\Gamma_2\) orders at approximately \(T_{\text{NF}_2} \sim 1.3\) K, and \(\Gamma_1\) at \(T_{\text{NF}_1} = 0.84\) K. At the lowest temperatures, the ratio between the two irreps stabilizes at 80%-20%.]

**D. 1D magnetic correlations in diffuse scattering**

Diffuse scattering can be harnessed to determine useful information, not only concerning the dimensionality of the interactions, but also the size and direction of the associated moments. Furthermore, it exposes the remaining magnetism from the sites which do not order.

The shape of the diffuse scattering is similar to what has been observed in \(\text{SrDy}_2\text{O}_4\) and \(\text{SrHo}_2\text{O}_4\). The sawtooth spectra isolated in Fig. 10 do not show the high frequency oscillations expected for the case of two- or threedimensional correlations\textsuperscript{8}. This is an indication that the fluctuations are mainly 1D. From our \textsc{multiX} calculations, we found that the moments are Ising-like. As these sites form zigzag chains along the \(c\)-axis, the simplest model representing these systems is the 1D ANNNI model\textsuperscript{18}. In its original iteration, the spins are collinear in a chain with two interactions: \(J_1\) between nearest neighbors and \(J_2\) between next-nearest neighbors. This model is equivalent to a zigzag chain where \(J_1\) is along the diagonal connection and \(J_2\) along the direction of the chain, which in our case is the \(c\)-axis.

For the analysis of the diffuse scattering, we calculated the partial differential neutron scattering cross-section...
per solid angle $\Omega$, per unit energy $E^4$:

$$\frac{d^2\sigma}{d\Omega dE} = \frac{|k|}{|k_i|} e^{-2W(Q)} \sum_{\alpha\beta} \left( \delta_{\alpha\beta} - \frac{Q\alpha Q\beta}{|Q|^2} \right) S^{\alpha\beta}_{\text{mag}}(Q, \omega),$$

where

$$S^{\alpha\beta}_{\text{mag}}(Q, \omega) = \left( \frac{\gamma n r_{0\beta}}{2} \right) \int dt e^{-i\omega t} \times \sum_{ll'} f'_l(Q) f'_{l'}(Q) e^{iQ(r_l - r_{l'})} \langle S^\alpha_{l}(0)S^\beta_{l'}(t) \rangle,$$

which is summed over the Cartesian coordinates $\alpha$ and $\beta$. Here, the scattering function $S^{\alpha\beta}_{\text{mag}}(Q, \omega)$ is defined as

$$S^{\alpha\beta}_{\text{mag}}(Q, \omega) = \left( \frac{\gamma n r_{0\beta}}{2} \right) \int dt e^{-i\omega t} \times \sum_{ll'} f'_l(Q) f'_{l'}(Q) e^{iQ(r_l - r_{l'})} \langle S^\alpha_{l}(0)S^\beta_{l'}(t) \rangle,$$

where $\langle S^\alpha_{l}(0)S^\beta_{l'}(t) \rangle$ is the spin-spin correlation function. Also, $\gamma_n$ is the gyromagnetic ratio of the neutron, $r_0$ the classical electron radius, $g$ the Landé g-factor, $k_i$ and $k_f$ the wave vectors of the incident and scattered neutrons, $Q = k_f - k_i$ the momentum transfer, $f(Q)$ the magnetic form factor, and $e^{-2W(Q)}$ the Debye-Waller factor. The sum has to be taken over all magnetic atoms. In an elastic case, where the energy transfer is negligible, a static approximation reduces the scattering function to

$$S^{\alpha\beta}_{\text{mag}}(Q) = \left( \frac{\gamma n r_{0\beta}}{2} \right) \times \sum_{ll'} f'_l(Q) f'_{l'}(Q) e^{iQ(r_l - r_{l'})} \langle S^\alpha_{l}(0)S^\beta_{l'}(t) \rangle.$$

The spin-spin correlation function can be written as

$$\langle S^\alpha_{l}(0)S^\beta_{l'}(t) \rangle = |\mu|^2 g_{ll'}(J_1, J_2, T),$$

where $g_{ll'}$ is the site dependent correlation function between the spins on positions $l$ and $l'$ for given interactions $J_1$ and $J_2$, at a temperature $T$. In the case of the 1D ANNl model, the correlation function is known in the thermodynamic limit. The powder average has to be calculated for comparison with the powder data. It is defined as the integral of the scattering function over the solid angle:

$$\bar{S}(Q) = \int \frac{d\Omega Q}{4\pi} S(Q).$$

The diffuse scattering is isolated from the diffraction pattern by subtracting the contributions from the nuclear and magnetic Bragg peaks. This results in the diffuse scattering intensity plus a background, as shown in Fig. 10. This figure shows the diffuse scattering for temperatures above and below the formation of order. Strong peaks originating from the copper sample holder were excluded from the spectra. This allows us to fit the residual spectrum using the magnetic interactions $J_1$ and $J_2$, and the moment size $|\mu|$, as adjustable parameters. The background is fitted with a smooth fourth order polynomial.

A closer inspection of the model shows that it is strongly dependent on the interaction ratio $J_2/J_1$ and is fairly insensitive to their individual values. Various combinations of $J_1$ and $J_2$, for a given interaction ratio, will produce a curve with a similar deviation $\chi^2$ between the model and the data. Unfortunately, this means that it is not possible to determine the absolute size of $J_1$ and $J_2$, but only their ratio $J_2/J_1$. However, the calculated spectra are sensitive to the sign of the interaction constants, i.e. whether they are ferromagnetic or antiferromagnetic. Note the sharp features in the diffuse scattering which are located near values of $|Q| = (n + \frac{1}{2})c^* = (2n + 1)\pi/c$. These positions are indicated by the vertical dashed lines in Fig. 10. For these sawtooth features, the sign of $J_1$ determines if the leading edge is to the left or the right of the dashed line. For a negative $J_1$, the edge is located on the right. That the periodicity is inversely proportional to $c$ is a further indication for the 1D character of these interactions.

Furthermore, the fact that neutrons interact only with magnetic moments which are perpendicular to the momentum transfer strongly affects the profile of the diffuse scattering. The dot product $Q \cdot (r_l - r_{l'})$ in $S^{\alpha\beta}_{\text{mag}}(Q, \omega)$ results in a much stronger contribution for any $Q$ along the rare earth chains, which run parallel to the $c$-axis. A spin orientation along $c$ produces a significantly lower scattering intensity with broader features than a spin in the $ab$-plane. For this reason, a spin along $c$ can be hard to detect in the presence of a large spin component residing in the $ab$-plane. The geometry of the chains also makes it hard to distinguish an alignment of the spins along $a$ or $b$.

The model contains one zigzag chain. In our struc-
ture, the two crystallographically inequivalent rare earth atoms are interacting along separated chains, resulting in a total of four chains within a chemical unit cell — two on site 1, two on site 2. As the two types of chain are sitting on two inequivalent sites, they are treated with their own set of magnetic interactions $J_1$ and $J_2$, and moment $\mu$. The only difference between the chains A and B of a given rare earth site will be the atomic position, as shown in Fig. 7. The total calculated spectra will then consist of the sum of the contributions from these four chains, plus a background. The number of atoms contained in a chain is set to such a high number, that the correlations converge to zero over the length of the chain.

While the model includes size and direction of the moments, only the sizes are fitted, with the directions fixed to the easy-axis obtained from the CEF calculations. The contributions from each site are shown as the orange and blue curves in Fig. 10. In order to keep the size of the moments stable, the polynomial background was fitted at a temperature where there is a clear and known contribution from both sites. The background was kept fixed afterwards. The size of the moments in Eq. 5 was scaled so that they match the values from the FULLPROF refinements. The results of this procedure are presented in Fig. 11.

Since the systems do not show significant structural change with temperature, the ratio of the exchange constants are therefore expected to remain constant. This ratio was obtained while simultaneously fitting several spectra recorded at a number of temperatures. This strategy was chosen to alleviate problems which could skew this value if too many spectra at temperatures close to the phase transition were chosen. Spectra taken at the lowest measured temperatures are also discarded, as different chains will eventually start to interact with each other.

1. BaDy$_2$O$_4$

The sharp sawtooth seen at $T = 1$ K suggests that only magnetism in the $ab$-plane is present, with no contribution from moments parallel to the $c$-axis, as this would result in broader features. This is also coherent with the direction of the moments obtained from multiX. The ANNNI model was then fitted with moments in the $ab$-plane for both sites. The experimental scattering intensity in Fig. 10(a)–(b) strongly decreases with temperature, but stabilizes at $T = 0.30$ K. It is interesting to note that the diffuse scattering intensity persists down to $T = 0.07$ K, after the onset of magnetic Bragg peaks. The diffusive intensity at $T = 0.07$ K still represented roughly half the size of the diffusive scattering intensity at $T = 1$ K. As expected, the trend of the magnetic scat-
tering intensity is the opposite to the one seen for the magnetic Bragg peaks — the intensity of the diffuse magnetic scattering decreases with lower temperatures. This seems to indicate that weight from the diffuse scattering is transferred to the magnetic Bragg peaks.

This leads to two possible scenarios presented in Fig. 11(a)–(b). In scenario (1), the size of the magnetic moments, which are responsible for the diffuse scattering on both sites, have the same temperature dependence, as seen in the thermal evolution of the magnetic Bragg peaks. However, this would mean that despite the magnetic order manifested through the presence of Bragg peaks, moments of the size of nearly 4μB and 2μB would continue to fluctuate down to \( T = 0.07 \) K on both sites. Or (2), a case where one of the site is not ordering. Here, the intensity of the diffuse scattering associated with site 2 is decreasing to zero for the lowest temperatures. The diffuse scattering seen at low temperature will then be entirely located on the site 1 which will remain in a strongly fluctuating state with a moment size of 5.2(2)μB. This is the scenario shown in Fig. 10(a)–(b), as the components are more easily separated, and the plot readable. For this case, the moment on site 1 was fitted at low temperatures, where only one contribution would be present, and kept fixed. This prevents that a similar contribution from the other site mixes in the fitting process.

For case (1), shown in Fig. 11(a), both sites have antiferromagnetic interactions with their nearest neighbors and next-nearest neighbors. The ratios \( J_2/J_1 \) are 0.59 and 0.60 for the site 1 and 2, respectively, which are both higher than 1/2. This implies that the atoms in the chains of both sites are interacting as a double Néel state, consistent with the FullProf analysis. For case (2), shown in Fig. 11(b), similar values are obtained with \( J_2/J_1 \) of 0.55 and 0.61 for the site 1 and 2, respectively. As expected, the site that does not order has a ratio \( J_2/J_1 \) which is closer to the critical value of 1/2.

The correlation function gives the distance over which sites are correlated, as shown in the lower panels of Fig. 10. Temperature and the ratio \( J_2/J_1 \) are affecting this length — it decreases when the temperature increases, and when the ratio \( J_2/J_1 \) is getting closer to 1/2. This is coherent with the observation that the ordered sites have larger ratio \( J_2/J_1 \) than the fluctuating sites, thus having correlations on a longer range. For case (1), where both sites remain in a fluctuating state, both will have an interaction range of about 20 neighbors at \( T = 1 \) K, which increases to nearly 150 neighbors at 0.20 K. For the case (2), where one site does not order, the correlation range is shorter at \( T = 1 \) K, with 10 neighbors, and it increases to 100 neighbors at 0.20 K.

Given that the two magnetic orders possess two different propagation vectors, it is likely to consider the two orders on separated sites. This was the interpretation resulting from the magnetic Bragg peaks refinement. Case (1) would then be the correct one. A proper interpretation was here not possible solely based on the diffuse scattering analysis.

2. \( \text{BaHo}_2\text{O}_4 \)

For \( \text{BaHo}_2\text{O}_4 \), the analysis of the diffuse scattering at \( T = 1 \) K is shown in Fig. 10(d). The shape of the spectra indicates the presence of magnetic moments in the \( ab \)-plane, as well as along \( c \)-axis. The component along \( c \), placed on site 2, decreases in intensity with temperature, as expected due to the magnetic order, which develops with moments along the \( c \) direction. The intensity of the component with the moment in the \( ab \)-plane, placed on site 1, remains constant in the temperature range from 2 K to 0.20 K, as shown in Fig. 11(c). This result suggests that site 1 does not order down to 0.20 K and remains in a strongly fluctuating state. From our analysis, we can thus conclude that the moment on site 1 lies in the \( ab \)-plane with a moment size of 8.8(3)μB, which is coherent with the multitX calculations. This is another indication for the fact that the magnetic order of both irreps coexist on site 2. The result of this fit is shown in Fig. 11(c), where the size of the moment on site 1 remains constant, and the one on site 2 decreases to zero. Here, one can clearly see the different transition temperatures of the two irreps on site 2. The intensity slightly decreases between \( T = 2 \) K and 0.80 K, as the order associated with \( \Gamma_2 \) develops, but then sharply drops when the order associated with \( \Gamma_1 \) sets in and the ordered moment becomes much larger.

There is no reason to believe that the diffuse scattering intensity on site 2 is not going to zero, but since this signal is broad and of low intensity at low temperature, it could be mistaken as part of the background. Therefore, it cannot be ruled out with certainty that site 2 is still slightly fluctuating.

Site 1, which does not order, is characterized by all antiferromagnetic interactions with a ratio \( J_2/J_1 = 0.56 \), similar to \( \text{BaDy}_2\text{O}_4 \)'s double Néel states. Site 2, which orders, has a much lower ratio of 0.33 — below the critical value of 1/2 — again with all antiferromagnetic interactions. It describes a simple Néel order, consistent with the FullProf analysis. The site that does not order also has a \( J_2/J_1 \) which is much closer to the critical value of 1/2. At \( T = 1 \) K, site 2 has correlations with up to 50 neighbors, while site 1 will interact with only 15 neighbors. At 0.20 K, the correlation length of site 1 increases to more than 150 sites.

The fits were limited to temperatures below 2 K, but for \( \text{BaHo}_2\text{O}_4 \), multiple spectra were recorded at temperatures above 2 K which show strong diffuse scattering and sharp features up to 60 K, as can be seen in Fig. 11(d). However, the ANNNI model with fixed \( J_1 \) and \( J_2 \) is unable to describe the observed spectra. A good fit would require unphysically large magnetic moment sizes — or a significant change in the values of \( J_1 \) and \( J_2 \), which is not expected, as there is no significant change in the crystal structure. On the other hand, it is possible that the higher temperatures are causing the population of higher CEF levels, which from the multitX calculation have been shown to be rather close in energy to the ground state. This could result in a set of more com-
Moments ($\mu_B$)

- **BaHo$_2$O$_4$:**
  - Case (1): $J_2/J_1 = 0.60$
  - Case (2): $J_2/J_1 = 0.59$
  - Case (3): $J_2/J_1 = 0.55$
  - Case (4): $J_2/J_1 = 0.61$

- **BaDy$_2$O$_4$:**
  - Case (1): $J_2/J_1 = 0.56$
  - Case (2): $J_2/J_1 = 0.57$

The position of the large features significantly and provides a better adjustment to the data, which would be required if the model would be applied at temperatures higher than 2 K. The calculations are also robust while cell parameters and rare earth positions are repositioned within the error bar — no change in the quality of the fit is noticeable, regardless of the temperature. The apparent shift in $J_2/J_1$ can thus not be explained by structural change.

Nonetheless, the quality of the fits at lower temperature is an indication for the presence of strong 1D correlations in these systems, as was observed in the case of SrDy$_2$O$_4$ and SrHo$_2$O$_4$. While additional interactions $J_3$ and $J_4$ between the chains, as defined in Fig. 7, are expected to be non-negligible at very low temperatures, the ANNNI model still adequately accounts for the diffusive part of our spectra.

### III. DISCUSSION AND CONCLUSION

In conclusion, we have performed CEF calculations using MULTIX to interpret our inelastic neutron scattering spectra. From these, we find a site-dependent magnetic anisotropy as well as the presence of an easy-axis, which is confined to either the ab-plane, or along the c-axis. We measured the temperature dependence of the neutron powder diffraction signal which we used to perform a refinement of the magnetic Bragg peaks. These spectra show strong diffusive components, which we fitted using a 1D ANNNI model, confirming the coexistence of long-range and short-range magnetic orders in both samples. In BaDy$_2$O$_4$, we observe magnetic order with two different wave vectors but a single transition temperature of $T_N = 0.48$ K. The ordered moments are restricted to the ab-plane. Magnetic fluctuations remain important to the lowest temperatures, even after the ordered moments saturate. The direction of the fluctuating moment is shown to be in the ab-plane, the same plane as the long-range order. For BaHo$_2$O$_4$, the magnetic order has two different irreps with the same propagation vector $k_0 = (0,0,0)$. The transitions occur at different temperatures, one at $T_{NR_1} = 0.84$ K, and the other at $T_{NR_2} \sim 1.3$ K. For both, the moments order along the c-axis. Magnetic fluctuations also remain after the order saturates, predominantly in the ab-plane. Agreements between the intensities of the modeled diffuse scattering and of the Bragg peaks indicate that interactions in these samples are for the most part 1D and confined within a specific zigzag chain.

This shares similarities with the behavior seen in Sr(Dy, Ho)$_2$O$_4$. In SrHo$_2$O$_4$, the sites 1 and 2 are host of a simple Néel long-range order along the c-axis, and a double Néel short-range order in the ab-plane, respectively. On the other hand, no long-range order has been found in SrDy$_2$O$_4$, but a detailed analysis of the diffuse scattering shows the presence of short-range correlations on both sites, with a spin alignment nonetheless in the same
The compounds SrHo$_2$O$_4$\textsuperscript{3,4–8}, SrEr$_2$O$_4$\textsuperscript{2,4}, and BaNd$_2$O$_4$\textsuperscript{48}, either through powders or single crystals neutron diffraction, are showing the coexistence of long-range (or quasi long-range) order and short-range order. So far, however, SrYb$_2$O$_4$\textsuperscript{12} is the only system where two long-range orders were reported. Other compounds such as BaTb$_2$O$_4$\textsuperscript{49} and SrDy$_2$O$_4$\textsuperscript{8} are only exhibiting short-range order, while BaTm$_2$O$_4$\textsuperscript{10} does not show any sign of ordering. Of all the compounds studied in this family, only SrGd$_2$O$_4$ shows two separated magnetic transition temperatures in zero-field, as seen from specific heat measurements\textsuperscript{50}. Its magnetic structure still remains unsolved. Another compound which stands out is SrTb$_2$O$_4$\textsuperscript{41} where the long-range order is incommensurate.

With our results on BaDy$_2$O$_4$ and BaHo$_2$O$_4$, we confirm the presence of two different types of long-range magnetic order — with different transition temperatures in one case — coexisting with fluctuating moments down to the lowest measured temperatures, unveiling two compounds with interesting and complex magnetic interactions. Measurement on single crystals will soon follow. This will allow us to describe more precisely the evolution of the diffuse scattering, especially with the use of polarized neutron scattering measurements which will help to separate the contributions from the different sites. This would also allow us to study the formation of magnetic domains which were seen in Sr(Dy,Ho)$_2$O$_4$\textsuperscript{7,10}.

A conservative scenario for both samples is that only one of the sites orders — hosting two types of magnetic order — and the other one remains fluctuating down to the lowest measured temperatures. However, from a combination of CEF calculations, refinement of the Bragg peaks, and a description of the nature of the diffuse scattering, there are strong indications for more exotic magnetic behaviors. In the case of BaDy$_2$O$_4$, we suggest the interesting possibility that both sites order, but keep fluctuating down to the lowest temperatures. This could then be a case of magnetic fragmentation, which as been observed in the spin liquid phase of a pyrochlore, where magnetic order and fluctuations coexist on the same site\textsuperscript{51,52}. Indications for a classical spin liquid ground state was indeed found from an investigation by ultrasound velocity measurements in the related compound SrDy$_2$O$_4$\textsuperscript{14}, as well as by $\mu$SR\textsuperscript{53}. BaDy$_2$O$_4$ might presents a new class of magnetic ground state whose nature is yet to be determined, which surely present an interesting challenge.

Concerning BaHo$_2$O$_4$, we propose a scenario in which only one of the two magnetic sites is ordering, initially at $T_{N\Gamma_2} \sim 1.3$ K. A second order on the same site, with a different irrep, takes place at $T_{N\Gamma_1} = 0.84$ K, leading to a separation of the magnetic phase. The two phase fractions evolve until they reach proportions of 20%–80%, where the fraction associated with $\Gamma_1$ becomes dominant. This scenario might be the result of the growing influence of the inter-chain interactions as the temperature is lowered. These interactions are required for long-range order, and their evolving influence could suddenly privilege one order over the other.

**ACKNOWLEDGMENTS**

The research at the Université de Montréal received support from the Natural Sciences and Engineering Research Council of Canada (NSERC), the Fonds de recherche du Québec Nature et technologies (FRQNT), and the Canada Research Chair Foundation, and the work at PSI received support from the Swiss National Foundation (SNF Grant No. 138018).
