

Competition between multiferroic and magnetic soliton lattice states in DyFeO₃

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(Received 30 April 2025; accepted 7 October 2025; published 17 November 2025)

Simultaneous breaking of time reversal and inversion symmetries in multiferroics couples ferroelectricity to magnetism and is a source of unusual physical phenomena that can be used in next-generation electronic devices. A notable example is DyFeO₃, which under applied magnetic fields exhibits a giant linear magnetoelectric response and a large spontaneous electric polarization induced by coexisting orders of Fe and Dy subsystems. Here, we use high-resolution neutron diffraction to show that at zero field DyFeO₃ hosts an incommensurate magnetic soliton lattice formed by spatially ordered Dy domain walls with an average domain size of 231(8) Å. The long-ranged interaction between the domain walls is mediated by magnons propagating through the Fe subsystem and is analogous to the Yukawa force in particle physics. An applied magnetic field destroys the long-ranged incommensurate order, unlocks the linear magnetoelectric response, and stabilizes the ferroelectric state. The magnetic domain walls are electrically charged and the soliton array dimerizes when both electric and magnetic fields are applied. Numerical simulations with experimental parameters suggest that the generic competition between the ferroelectric and incommensurate states can be effectively controlled by an applied electric field.

DOI: [10.1103/kwrl-x4hw](https://doi.org/10.1103/kwrl-x4hw)

I. INTRODUCTION

Multiferroics are intriguing materials that combine ferroelectric and magnetic ordering, presenting exciting opportunities for advanced applications in memory devices, sensors, and spintronics [1–4]. In many multiferroics, the electric polarization arises from two coexisting magnetic orders—one even and one odd under inversion symmetry [4]. The symmetry also permits a free energy term known as the Lifshitz invariant, which favors a state with periodically modulated order parameters. Often, these modulations are incommensurate (IC) with the crystal lattice, creating a competition with the uniform ferroelectric state [4]. Thus, investigating materials with coexisting order parameters is essential to advance our understanding of multiferroism.

Rare-earth orthoferrites RFeO₃ host two interpenetrating magnetic sublattices, which attracts persistent attention over the last decades due to unusual physical properties induced by the interaction between them. They exhibit a spontaneous spin-reorientation (SR) transition, [5,6], ultrafast laser-induced magnetization switching [7,8], multimagnon

spin excitations [9], and multiferroicity [10,11]. Magnetism in these compounds is determined by strongly hierarchical interactions. The behavior of the Fe subsystem is primarily governed by the Fe-Fe exchange coupling with a simple G-type antiferromagnetic (AFM) order ($\mathbf{k} = 0$) and a much smaller Dzyaloshinskii-Moriya interaction gives rise to canted spins and hence a weak ferromagnetic (FM) moment [12]. A weak magnetic anisotropy orients the Néel vector $\mathbf{n} \parallel c$ axis and the resulting magnetic ground state is described by a Γ_4 irreducible representation (irrep). With decreasing temperature, some orthoferrites show an SR from Γ_4 to Γ_2 or Γ_1 phases [5]. At even lower temperatures, typically $T < 10$ K, the rare-earth subsystem in some orthoferrites orders antiferromagnetically and the type of AFM ordering is primarily determined by a crystal electric field (CEF)-induced anisotropy. More specifically, it varies between a collinear c -axis structure (CeFeO₃ [13] and ErFeO₃ [14]) or a non-collinear order when the rare-earth moments lie within the ab plane (TbFeO₃ [15], HoFeO₃ [16], and DyFeO₃ [17,18]).

In rare-earth orthoferrites with $R = \text{Gd, Tb, and Dy}$, antiferromagnetically ordered Fe spins preserve the inversion symmetry of the crystal lattice, whereas a rare-earth spin ordering breaks it [4]. The coexistence of two different orders is made possible by the frustration of interactions between Fe and R spins, forming two interpenetrating cubic sublattices. The frustration is lifted either by a polar shift of the R ions with respect to Fe ions, which leads to ferroelectricity, or by an incommensurate modulation of the two magnetic orders. GdFeO₃ becomes ferroelectric below the Gd ordering

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temperature [11], whereas TbFeO₃ shows an IC soliton array state that involves both Fe and Tb spins at an applied magnetic field [15]. The competition between the ferroelectric and IC states in orthoferrites is not accidental: when magnetoelectric coupling $P\Gamma\Gamma'$ is allowed by symmetry, so is the Lifshitz invariant, $\Gamma \overleftrightarrow{\partial} \Gamma' = \Gamma \partial \Gamma' - \Gamma' \partial \Gamma$, ∂ being the derivative along the polarization direction, which favors periodically modulated magnetic orders in *centrosymmetric* magnets.

In this work, we study the competition of IC order and the ferroelectric state in DyFeO₃ using a combined approach of single-crystal neutron diffraction and theory analysis. We show that at zero field Dy domain walls (DWs) form a periodic array stabilized by long-ranged interactions mediated by a magnon exchange, similar to that observed in TbFeO₃. The application of a magnetic field along the c axis suppresses the IC state in favor of a ferroelectric state with short-range commensurate order at $B \approx 2.6$ T. Due to the magnetoelectric coupling, the IC state can also be suppressed by applying an electric field. For zero magnetic field the critical electric field is rather high, but it decreases fast under applied magnetic fields, making the electric control of magnetism in DyFeO₃ feasible.

II. EXPERIMENTAL DETAILS

The single crystal of DyFeO₃ was grown by the floating-zone method using an optical floating-zone furnace (FZ-T-10000-H-IV-VP-PC, Crystal System Corp., Japan) with four 1000 W halogen lamps as a heat source as described in Ref. [18]. DyFeO₃ crystallizes in $Pbnm$ space group with low-temperature lattice parameters, $a = 5.293$ Å, $b = 5.584$ Å, and $c = 7.587$ Å. For the neutron scattering experiments, we used a DyFeO₃ crystal with a mass of $m = 19$ mg [18].

We performed three neutron diffraction measurements using the cold-neutron triple-axis spectrometer (TASP) at SINQ, PSI. In the zero-field experiment, the sample was oriented in the $(h, 0, l)$ scattering plane and installed in the orange cryostat. For the first in-field measurements, we oriented the crystal in the (h, h, l) scattering plane and a horizontal cryomagnet MA-7 was used to produce magnetic field. In the second in-field experiment, we oriented the sample in the $(h, 0, l)$ plane and applied a vertical field along the b axis using the MA-10 cryomagnet. The base temperature in all experiments was ≈ 1.7 K. We operated the instrument in the diffraction mode (no analyzer used) with $k_i = 1.3$ Å⁻¹. Two 40 arc min collimators were installed before and after the sample. A cold Be filter was used to suppress a higher-order harmonic contribution.

III. RESULTS

A. Single-ion physics of Dy³⁺ and the magnetic structure of DyFeO₃

DyFeO₃ hosts two interpenetrating magnetic sublattices: The Fe sublattice orders into a Γ_4 state at $T_N \approx 600$ K and exhibits a reorientation transition toward a Γ_1 state at $T_{SR} = 73$ K, while the Dy moment orders below 4 K [5,17]. However, there was controversy regarding the Dy order: single-crystal neutron diffraction experiments yielded a commensurate order of Dy moments with $\mathbf{k}_m = (0, 0, 1)$ (Γ_5 irrep)

[19]. Interestingly, the symmetry of the Dy order allows for a $\Gamma_4 \overleftrightarrow{\partial} \Gamma_5$ Lifshitz invariant that favors an IC modulation along the c axis, which could suppress an electric polarization. Recent high-resolution powder diffraction experiments observed a small splitting of the $(0, 0, 1)$ peak suggesting the presence of IC modulations along the c axis, $\mathbf{k}_m = (0, 0, 1 \pm \delta)$ [17,18]. A refinement of the powder diffraction data did not allow to uniquely identify the ordering pattern of Dy spins suggesting either a nonharmonic spin-density wave (SDW) order or an elliptical spiral as the ground state, yet there is no single-crystal data supporting this conclusion raising the question whether the IC order is realized exclusively in powder samples [17].

The type of AFM ordering of the R ions in RMO_3 is primarily determined by the (CEF-induced anisotropy. In the isostructural material DyScO₃, point-charge model (PCM) calculations supported by experimental data demonstrated that Dy moments have a very strong Ising-like anisotropy with the quantization axis in the ab plane, $\approx \pm 28^\circ$ from the b axis [21,22]. We performed an analysis of the single-ion ground state of Dy³⁺ ions using PCM and found that the anisotropy in DyFeO₃ is very similar to DyScO₃ (see details in Appendix A). The calculated canting angle is close, $\approx \pm 29^\circ$ [see Fig. 1(e)]. The ground state doublet is $|\psi_\pm\rangle \approx |\pm 15/2\rangle$, and it has only a negligible ($< 1\%$) admixture of other states. The first excited state primarily consists of $|\pm 13/2\rangle$ states located at 28 meV, making the contribution of the excited states to low-temperature properties below 10 K, negligible.

This result is important in the context of magnetic structure determination. The PCM calculations yield a strongly anisotropic Ising-like nature of Dy moments, making the spiral solution implausible because it involves a continuous rotation of the Dy moments within the ab plane. The second solution yields two SDW with nonequivalent polarization directions. Both lie within the ab plane, rotated by $\pm 29^\circ$ with respect to the b axis. This polarization direction perfectly coincides with the direction of the Ising axis obtained from the PCM calculations, providing strong support for the SDW-like scenario. We, however, note that the general-type SDW involving a continuous modulation of the moment size is also unlikely for the Ising doublet with $\psi_\pm = |\pm 15/2\rangle$. Instead, the incommensurability in the Ising-class models (such as an anisotropic next-nearest neighbor Ising model) can arise from the spatial ordering of the DW [23] similar to one observed in TbFeO₃ [15].

B. Magnetic structure studied by neutron scattering

We studied the field-induced evolution of magnetic order in DyFeO₃ by means of high-resolution single-crystal neutron diffraction using the TASP instrument at PSI. We focused primarily on the close vicinity of the $(0, 0, 1)$ peak [24]. This reflection is associated with AFM order along c axis within the rare-earth subsystem as was found in isostructural compounds DyScO₃ [21,22] and YbAlO₃ [25,26] and is the strongest magnetic reflection of the Dy subsystem [17]. Figure 1(c) shows the representative diffraction profiles measured at zero field and base temperature. It exhibits two strong incommensurate peaks at $\mathbf{k}_m = (0, 0, 1 \pm \delta)$, four additional satellites at

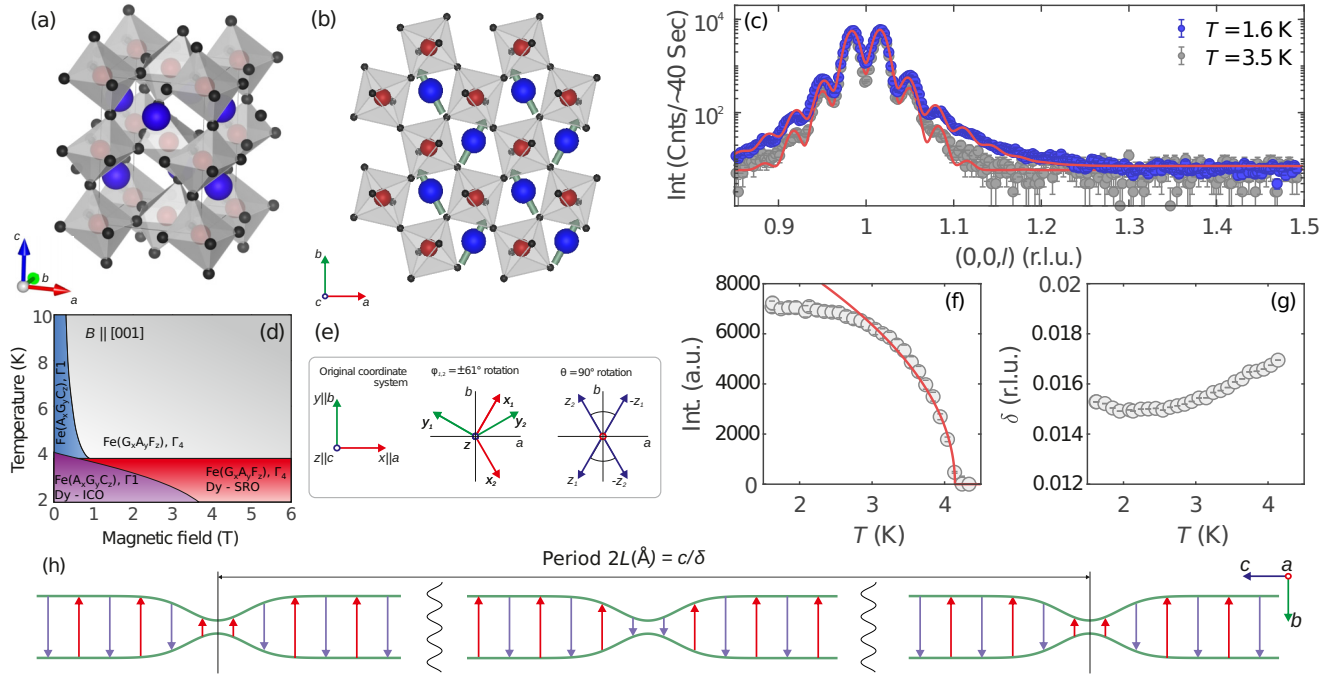


FIG. 1. Formation of magnetic soliton lattice in DyFeO_3 . (a), (b) crystal structure of DyFeO_3 . Blue, red, and black balls represent Dy, Fe, and O ions, respectively. Green arrows in panel (b) demonstrate the ordering pattern of Dy moments in the ab layer at zero field. (c) Neutron diffraction data measured along the $(0, 0, l)$ direction at $T = 1.7$ and 3.5 K. The intensity is shown on a logarithmic scale. Red lines show the fitting results with the Monte Carlo model as described in the main text. The absorption correction was included in the calculated curve. (d) Schematic field-temperature phase diagram of DyFeO_3 [19] for $B \parallel c$. The blue and gray areas correspond to Γ_1 and Γ_4 magnetic orders of the Fe subsystem, and the disordered state within the Dy subsystem. The violet area corresponds to the IC Dy order. The application of a magnetic field causes a spin reorientation of Fe moments, $\Gamma_1 \rightarrow \Gamma_4$ and stabilizes a short-range commensurate Dy order (red area). (e) Visualization of the Ising axis directions of Dy^{3+} moments. The axes lie within the ab plane at $\pm 29^\circ$ to the b axis (see Appendix A). (f). (g) Temperature dependence of intensity (f) and IC parameter (g) of the first harmonics of Dy order (see Appendix C for details). (h) Sketch of the incommensurate magnetic structure of the Dy subsystem. Dy moments form an AFM structure with a domain size $L = \frac{c}{2\delta}$.

$3\mathbf{k}_m$ and $5\mathbf{k}_m$, and broad diffuse scattering extending up to ≈ 1.3 r.l.u. The even-order satellites at $2n\mathbf{k}_m$ have not been observed at zero field [27]. The IC parameter $\delta = 0.01646(8)$ slightly differs from the value obtained from powder measurements by Ritter *et al.* $\delta = 0.026$ – 0.028 [17] but agrees nicely with $\delta = 0.0173(5)$ obtained by Biswas *et al.* [18]. Such a difference indicates a sample dependence of the propagation wave vector. δ allows us to directly estimate the real space periodicity of the magnetic structure, $c/\delta = 462(16)$ Å. With increasing temperature, the intensity of the AFM peak is continuously suppressed, eventually disappearing at $T_N = 4.14(2)$ K [see Fig. 1(f)]. The IC parameter δ exhibits a minor increase between 1.6 K and T_N [Fig. 1(g)].

Next, we looked into the field-induced evolution of the magnetic order. When a magnetic field is applied along the easy axis, $B \parallel b$, it quickly polarizes Dy moments and stabilizes a field-induced FM state, as evident from a sharp jump and quick saturation of the magnetization at $B \geq 0.5$ T [Fig. 2(c)]. Neutron data yield a strong suppression of the AFM reflections at the same magnetic field, as demonstrated in Figs. 2(a) and 2(c). The parameter δ exhibits a moderate enhancement with field (we discuss the possible reasons of this effect in Sec. III D) and shows hysteretic behavior, likely related to the DW pinning. Additional data measured for another in-plane

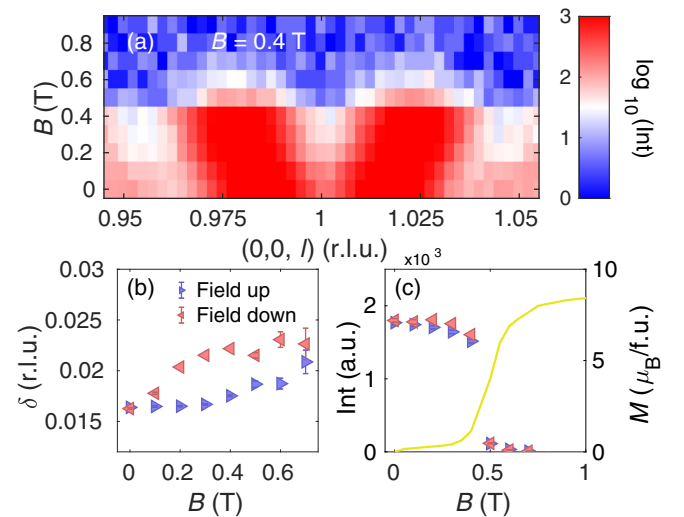


FIG. 2. (a) Field dependence of the neutron intensity measured along $(0, 0, l)$ at $T = 1.7$ K with the magnetic field applied along the b axis (easy axis of magnetization). The intensity is shown on a logarithmic scale. (b), (c) Field dependence of the δ parameter (b) and peak intensity (c) obtained with the magnetic field ramped up (blue) and down (red). Yellow line in panel (c) demonstrates bulk magnetization from Ref. [20] measured at $T = 2$ K.

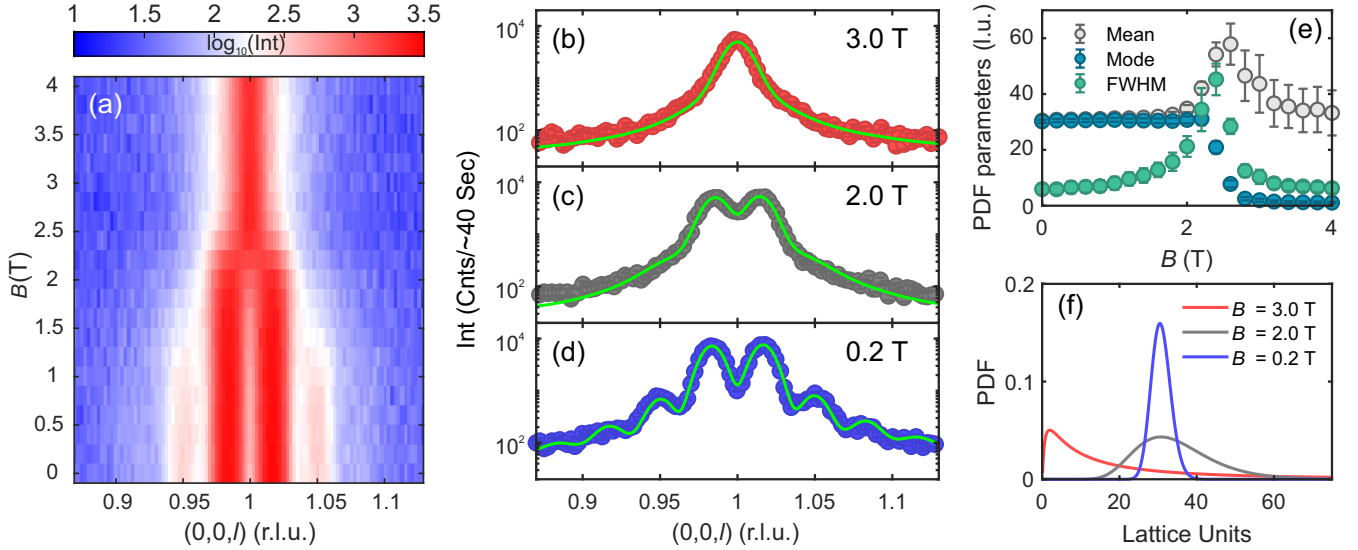


FIG. 3. Field-induced suppression of the soliton lattice and domain wall distribution. (a) Field dependence of the neutron intensity measured along $(0, 0, l)$ at $T = 1.7$ K. The intensity is shown on a logarithmic scale with the magnetic field applied along the c axis. Representative scans along the $(0, 0, l)$ direction taken at 3 T (b), 2 T (c), and 0.2 T (d) (dots represent the measured signal). The green lines are the absorption corrected fits to the data using the Monte Carlo model with the intensity shown on a logarithmic scale. (e) Mode, mean, and full width half maximum (FWHM) of the PDF fitted for different fields. (f) Representative PDF extracted from our model of the data shown in panels (b)–(d).

field geometry, $B \parallel [110]$, demonstrate qualitatively similar behavior and are summarized in Appendix B.

The magnetic field applied along the c axis causes very different effect as can be seen in Fig. 3(a). We can broadly categorize the field-induced evolution into three regimes: (1) at $B < 2.2$ T, the positions of the primary peaks remain constant, but the higher-order satellites continuously decrease in intensity forming a broad diffusive tail; (2) at $2.4 < B < 2.6$ T, two primary IC satellites merge into a commensurate peak at $(0, 0, 1)$; (3) at $B > 2.6$ T, a single Lorentzian peak centered at $(0, 0, 1)$ dominates the pattern. Three representative patterns taken at 3, 2, and 0.2 T are shown in Figs. 3(b)–3(d).

Thus, our single crystal data clearly support the formation of the IC state observed in powder diffraction experiment at zero field [17,18]. A magnetic field applied along an in-plane direction directly couples to the Ising-like Dy moments and quickly polarizes the Dy subsystem as shown in Fig. 2 for $B \parallel b$ and in Sec. B for $B \parallel [110]$. In contrast, a magnetic field applied along the c axis (hard axis of Dy moments) primarily interacts with the Fe subsystem, causing the $\Gamma_1 \rightarrow \Gamma_4$ transition. In addition, $B \parallel c$ indirectly controls Dy order via the Dy-Fe coupling. As we show next, the AFM order of Dy spins does not change locally, but rather suppresses the IC long-range ordering of the domain walls in favor of the commensurate AFM state with randomly positioned domain walls. Ferroelectricity and a strong linear magnetoelectric response are observed in the commensurate phase [10].

C. Monte Carlo simulations of neutron diffraction data

In view of the Ising nature of Dy moments, the most plausible picture of the IC state is the spatial ordering of Dy domain walls similar to the one observed in TbFeO_3 [15].

These walls separate the domains with opposite signs of the AFM Dy moments order and form a magnetic soliton lattice [Fig. 1(h)]. Following this idea, we describe our neutron scattering data using an empirical Monte Carlo approach. We consider an AFM Ising spin chain as schematically demonstrated in Fig. 1(h) and assume that the distances between domain walls (i.e., domain sizes) follow a log-normal distribution. The probability density function (PDF) for a log-norm distribution can be written as follows:

$$P(x) = \frac{1}{x\sigma\sqrt{2\pi}} \exp\left(-\frac{(\ln x - \mu)^2}{2\sigma^2}\right), \quad (1)$$

where parameters σ and μ are the logarithm of scale and the logarithm of location, respectively. Since the distribution is not symmetrical, the mode (the most probable value) and the mean are not the same and are defined as $\exp(\mu - \sigma^2)$ and as $\exp(\mu + \sigma^2/2)$, respectively. The log-normal FWHM is given by $\exp[(\mu - \sigma^2) + \sqrt{2\sigma^2 \ln 2}] - \exp[(\mu - \sigma^2) - \sqrt{2\sigma^2 \ln 2}]$.

Using this approach, we can sample large chains with domain configurations where the randomness is controlled by the parameters of the log-normal PDF (see details in Sec. D). The diffraction pattern calculated for the fully ordered domain wall pattern with a fixed distance between the defects is very distinctive [Fig. 4(a1)]. The higher harmonics show no intrinsic width, with only instrumental broadening being present. The intensity of the higher-order satellites decays much slower compared to the experiment. These discrepancies arise due to the partial disorder in the domain configuration of DyFeO_3 . The introduction of partial disorder leads to a broadening of the higher-order harmonics, bringing the simulated pattern much closer to the experimental

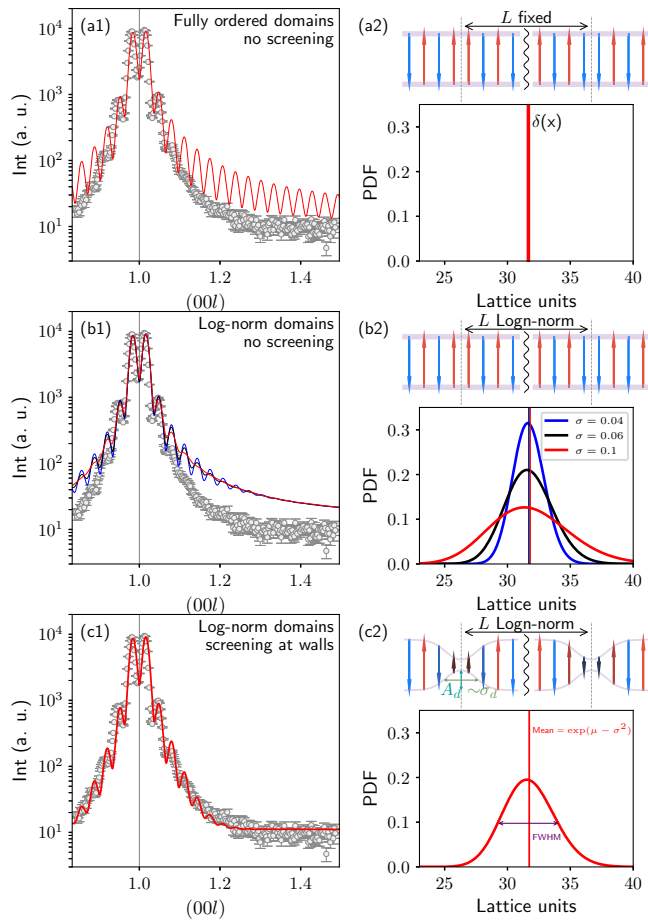


FIG. 4. Monte Carlo simulations. The left-side panels are diffraction patterns: The blue points are neutron diffraction at 1.5 K in zero field, the solid lines are the simulated patterns. The right-side panels show the type of used domain walls and PDF. (a1), (a2) The case of fully ordered domains with abrupt domain walls. (b1), (b2) The partially disordered defects, abrupt domain walls. (c1), (c2) The partially disordered defects, “screened” domain walls.

data [Fig. 4(b1)]. However, the intensity in the experimental data decays much faster with increasing $|\mathbf{Q}|$. The model overestimates the intensity of the tails, and we found that this discrepancy cannot be resolved by adjusting the PDF parameters. The parameter σ controls the smearing of the peaks but has no effect on the $|\mathbf{Q}|$ -dependence of the intensity decay. Furthermore, the parameter μ is constrained by the propagation vector, which must align with the mean of the distribution. We also considered several other PDF functions (Gaussian, Lorentzian, hat, box), but it still did not allow us to reduce the high- $|\mathbf{Q}|$ intensity. Therefore, additional degrees of freedom are needed in the model to accurately reproduce the diffraction pattern of DyFeO_3 .

Instead of *sharp*, abrupt defects, the domain walls are often smooth and have a finite size. In the presence of extreme anisotropy, the Ising spins have only one degree of freedom—the magnitude of the magnetic moment—which can continuously change across the defect. In this scenario, the magnetic moments have a reduced magnitude at the defect location, gradually regaining their nominal value deeper within the domain [Fig. 4(c2)]. The screening can be included

into the model by multiplying the magnetic moments by a reduction factor, which follows a Gaussian function taking the distance from the domain wall as argument. This Gaussian screening has two parameters: An amplitude A_s , which dictates how strong the screening is, and σ_s , which defines the extent of the screening (or, equivalently, the domain wall size). The top panel in Fig. 4(2c) illustrates how the screening compares to the *sharp* domain walls [Figs. 4(a2) and (b2)]. As will be shown in Sec. III D, the domain walls carry electric charge and therefore produce local distortion of the crystal structure close to the domain wall position. This distortion will modify the CEF Hamiltonian and magnetic anisotropy. We speculate that this effect can be responsible for the reduction of Dy moment at the domain wall due to deviation of the ground-state wavefunction from the pure state with $\psi_{\pm} = |\pm 15/2\rangle$.

The model quantitatively reproduces the observed data, both at zero and finite field [see Figs. 3(b)–3(d) and 1(c)]. The fit of the zero field [Fig. 1(c)] data at 1.6 K yields that an average distance between domain walls is 30.4(1.0) l.u. [231(8) Å] and the distribution function is reasonably sharp with an FWHM of 6(2) l.u. indicating that the domain size is well defined. The presence of a long diffusive tails in the data helps to extract details on the fine structure of the domain walls: the maximal amplitude of the moment suppression is 58(6)% and the FWHM of the domain wall is 3.1(2) l.u.

As the next step, we fitted the data collected with $B \parallel [001]$ using the same approach, Figs. 3(b)–3(d). The field dependence of mean, mode (the most likely value), and FWHM of the fitted distribution is summarized in Fig. 3(e). In the low-field regime, the mode and mean coincide and remain roughly constant up to ≈ 2 T, while the FWHM continuously grows. This increase reflects a continuous broadening of the PDF and corresponds to the suppression of higher-order harmonics at $3\mathbf{k}_m$ and $5\mathbf{k}_m$. Above 2.2 T, the mode rapidly decreases, while the mean, showing a moderate increase at intermediate fields, eventually returns to the initial low-field value. The fitted PDF for three representative fields are summarized in Fig. 3(f). One can see a sharp peak at 0.2 T, which broadens considerably at 2 T yet remains at the same position. At higher fields, the peak shifts toward zero reflecting the formation of the short-range ordered state with a finite correlation length determined by the value of the mean.

Our Monte Carlo analysis yields formation of the soliton lattice state with relatively low degree of disorder (variance of the domain size) at low fields. Application of low fields does not change the average distance between the domain walls, but continuously increases the disorder (domain size variance). At $B > 2.6$ T (above the $\Gamma 1 \rightarrow \Gamma 4$ transition), the incommensurate order is suppressed in favor of the commensurate state. This state has finite correlation length due to the presence of domain wall defects and our results yield that the average distance between the defects is similar in both, low- and high-field states [gray curve in Fig. 3(e)].

D. Theoretical model

To rationalize the field-induced evolution of the magnetic state, we consider a model in which nearly isotropic Fe spins are described by the Néel vector $\mathbf{n} = (\cos(\phi), \sin(\phi), 0)$. For

$H\|c$, $\mathbf{n}$ stays in the ab plane, and the AFM ordering of strongly anisotropic Dy spins is described by the Ising variable $\sigma = \pm 1$. The free energy density of the IC state in zero fields is

$$f = \frac{J}{2} \left(\frac{d\phi}{dz} \right)^2 + U_a(\phi) + \sum_n (f_{\text{Dy}}^{(0)} - 2gQ_n n_x(z)) \delta(z - z_n), \quad (2)$$

where the first two terms are the exchange and anisotropy of the Fe subsystem. $f_{\text{Dy}}^{(0)}$ is the bare free energy of the Dy DW with a topological charge $Q_n = \frac{1}{2}[\sigma(z_n + 0) - \sigma(z_n - 0)] = (-1)^n$ located at $z = z_n$. The last term results from the Lifshitz invariant, $\Gamma_4 \overleftrightarrow{\partial}_z \Gamma_5$, describing the interaction between Fe and Dy moments at DWs. This interaction stabilizes an IC modulation along the c axis, in contrast to TbFeO_3 , where the $\Gamma_8 \overleftrightarrow{\partial}_y \Gamma_2$ invariant produced an IC modulation along the b axis, $\mathbf{k}_m = (0, \pm\delta, 1)$. Details of calculations and parameter choice can be found in Appendix E.

The stabilization of the IC state has a transparent physical interpretation in the weak coupling limit, when the deviation of ϕ from 90° is relatively small. In this case, ϕ plays the role of a massive scalar field mediating interactions between Dy domain walls similar to the Yukawa interaction between nucleons resulting from pion exchange [28]. The interaction between two patches of domain walls with topological charges Q_1 and Q_2 , separated by a distance r , is the screened Coulomb interaction (with opposite sign) $\propto -Q_1 Q_2 \frac{1}{r} e^{-r/l}$, where the screening length l is inversely proportional to the gap in the magnon spectrum propagating through the Fe magnetic subsystem. The “dressing” of the Dy domain wall by distortions of the Fe ordering decreases its energy:

$$f_{\text{Dy}} = f_{\text{Dy}}^{(0)} \left(1 - \frac{g^2}{f_{\text{Dy}}^{(0)} \sqrt{JK_x}} \right), \quad (3)$$

where K_x is the strength of the second-order magnetic anisotropy. Above a critical value of the coupling constant g , the domain wall free energy becomes negative and the IC state appears. Neighboring domain walls have opposite topological charges and repel each other, which keeps them at a distance $\propto l$ and determines the IC parameter δ . The IC state suppresses the uniform FE state, which explains why electric polarization was not observed at low magnetic fields [10].

What makes DyFeO_3 different from TbFeO_3 is the electric and magnetic moments induced by the coexisting Fe and Dy orders:

$$\begin{aligned} P_z &= \sigma P_0 \cos \phi + \alpha \sigma H_z, \\ M_z &= M_{\text{WFM}} \cos \phi + \alpha \sigma E_z, \end{aligned} \quad (4)$$

where $P_0 = 1.5 \times 10^3 \mu\text{C m}^{-2}$ is the spontaneous electric polarization, $M_{\text{WFM}} = 0.13 \mu_B$ is the weak FM moment per Fe ion induced along the c axis by $n_x = \cos \phi$ and α is the linear magnetoelectric coefficient in the uniform Dy state with a Γ_5 symmetry. Due to depolarization and demagnetization effects, the electric and magnetic fields, E_z and H_z , in Eq. (4) differ from the applied fields, E_0 and H_0 . Whereas the weak

demagnetization field can be neglected, the depolarization field $\sim 70 \text{ kV cm}^{-1}$ effectively modifies the anisotropy of Fe spins and their weak FM moment in the IC state (see Appendix E2).

Figures 5(c) and 5(d) show the z dependence of ϕ and σ in the IC state at $H_0 = E_0 = 0$, in which Dy domain walls form an equidistant array and ϕ describes the direction of the Néel vector oscillating around the average value of 90° (dashed line). Figure 5(e) shows the corresponding variation of the electric polarization. Negatively charged domain walls (the electric charge density, $\rho = -\frac{dP_z}{dz}$) are surrounded by a cloud of compensating positive charges. For $H_0 \neq 0, E_0 = 0$, as well as for $H_0 = 0, E_0 \neq 0$, the domain wall array remains equidistant and has a zero net polarization. The alternation of the sign of σ suppresses the linear magnetoelectric response. However, when both H_0 and E_0 are nonzero, the array is dimerized [see Figs. 5(f) and 5(g)] with P parallel (antiparallel) to the applied electric field in longer (shorter) σ domains. The dimerization results from the linear magnetoelectric effect.

Figure 5(a) shows the DyFeO_3 phase diagram in electric and magnetic fields applied along the c axis, obtained by numerical minimization of the free energy using parameters taken from the experiment. Under $H\|c$, the weak FM moment induced by $n\|a$ forces the Néel vector in the IC state to tilt (on average) toward the a axis. The decrease of the average angle, ϕ_* , [dashed line in Fig. 5(c)] effectively reduces the Fe-Dy coupling, $g \rightarrow g \sin \phi_*$, which increases the energy of the IC state. At a critical field, H_{c1} , the IC state (blue color) undergoes a first-order transition into the uniform FE_1 state (gray), in which both Dy and Fe order parameters are uniform, but the Néel vector is not yet fully rotated to the a axis. In the FE_2 state (red), $\phi = 0$. The transition between the FE_1 and FE_2 states is abrupt for a sufficiently strong fourth-order anisotropy: $U_a(\phi) = \frac{K_x}{2} \cos^2 \phi + \frac{K'_x}{4} \cos^4 \phi$, with $K'_x < 0$.

The calculated magnetic field dependence of the electric polarization, $P(H)$, shows a very good quantitative agreement with the experimental data by Tokunaga *et al.* [10]. However, while our theory predicts the formation of a uniform FE state above H_{c1} , the experimentally observed order exhibits a large, but finite correlation length [250(60) Å at 4 T, see Fig. 3(b)], related to the metastability of the DW array and pinning of domain walls by disorder. The domain wall pinning can further explain the minor difference between the propagation wave vector in our samples and in Ref. [17].

The behavior of $H\|b$ can be understood based on the fact that domain walls in the Ising-like A_y -ordering of Dy spins carry an FM moment (anti)parallel to the b axis. A small adjustment of the domain wall positions therefore aligns the magnetic moments of all domain walls along the magnetic field direction, which reduces the domain wall free energy: $f_{\text{Dy}}^{(0)} \rightarrow f_{\text{Dy}}^{(0)} - \cos \alpha_{\text{Dy}} \mu_{\text{Dy}} H_b$, where $\mu_{\text{Dy}} = 10 \mu_B$ is the Dy magnetic moment and α_{Dy} is the angle between the spin-quantization axis of Dy ions and the b axis. The result is the decrease of the period of the IC state for $H\|b$, until the uniform $F_y C_x$ -state sets in (see Fig. 2).

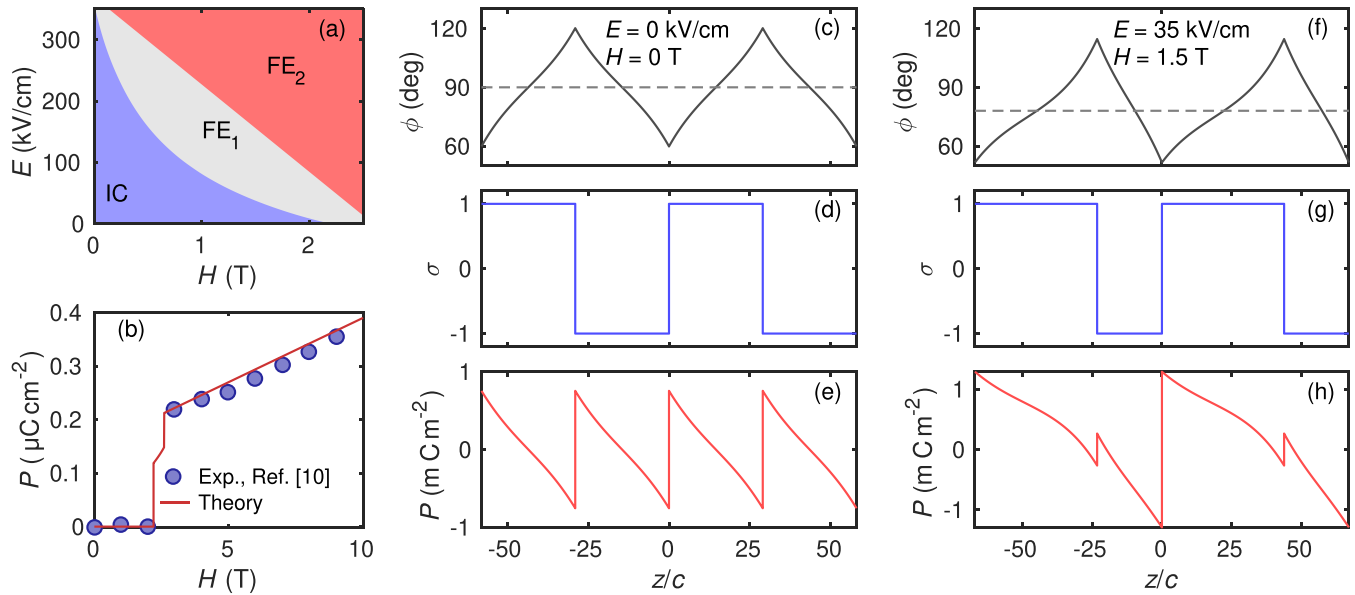


FIG. 5. Origin of the incommensurate order and the phase diagram of DyFeO_3 . (a) Three phases obtained by numerical minimization of the free energy density in applied electric and magnetic fields: the IC state (blue), the uniform ferroelectric state, FE_1 with $\sigma = 1$ and $0 < \phi < 90^\circ$ (gray), and the uniform ferroelectric state FE_2 (red) with Dy moments in the Γ_5 state and $\phi = 0$ (Γ_4 symmetry). (b) Magnetic field dependence of the electric polarization from Ref. [10] and calculated by our theory. (c)–(h) Coordinate dependence of the angle ϕ describing the direction of the Néel vector in the IC state (c), (f), the sign change of the Dy order parameter σ (d), (g), and the electric polarization (e), (h). Data are calculated for $H_0 = E_0 = 0$ (c)–(e) and $H_0 = 1.5$ T, $E_0 = 35$ kV cm $^{-1}$ (f)–(h). The dashed line in the panels (c), (f) shows the average value of ϕ . Distances are measured in units of the lattice constant c .

IV. DISCUSSION AND CONCLUSION

Frustration due to interactions between rare earth and Fe spins in orthoferrites can be lifted in two different ways: by polar lattice distortions inducing an electric polarization or by periodic modulations of the two magnetic orders, which gives rise to the competition between uniform multiferroic and IC states. In this work, we used a combination of high-resolution neutron diffraction, Monte Carlo simulations, and model calculations to understand the complex low-temperature behavior of DyFeO_3 . We showed that in low fields, DWs in the AFM ordering of Dy spins form a periodic array (soliton lattice) stabilized by the interactions with Fe spins at the DWs. Due to long-ranged interactions mediated by magnons propagating through the Fe magnetic sublattice, the sharp Dy domain walls effectively acquire a large width, comparable to the width of domain walls in the AFM ordering of Fe spins, which explains the long period of the IC state. The IC state is the ground state of the system, but it can be suppressed by the application of magnetic and/or electric fields. A magnetic field applied within the ab -plane couples directly to Dy moments, reduces the domain size and eventually stabilizes field-induced FM state. In contrast, the interaction of the c -axis magnetic field with Dy spins lying within the ab plane is weak. Instead, it causes the re-orientation of Fe spins toward the a axis, which suppresses the IC state and induces a uniform AFM order of Dy spins with a finite correlation length, presumably due to remaining randomly positioned domain walls. This state has a spontaneous electric polarization due to polar shifts of Dy ions and shows a giant linear magnetoelectric effect observed in Ref. [10].

The linear magnetoelectric coupling allows to control the magnetic soliton lattice: When both electric and magnetic fields are applied along the c axis, the IC state dimerizes—the widths of neighboring domains become unequal [Figs. 5(d) and 5(g)]. We estimate that in zero field, the IC state is stable up to high electric fields of ~ 350 kV cm $^{-1}$. However, when a magnetic field is applied, the critical electric field is strongly reduced and comes down to the field range used in the study of electric polarization of DyFeO_3 [10]: $E = 80$ kV cm $^{-1}$ at $\mu_0 H_0 = 1$ T and $E = 35$ kV cm $^{-1}$ at $\mu_0 H_0 = 1.5$ T [see Fig. 5(a)]. It is still a question whether the Dy domain walls can be moved and annihilated at low temperatures. However, our calculations show that the electric control of magnetism in DyFeO_3 might be possible close to H_{c1} .

Our analysis indicates that the ground state of DyFeO_3 is determined by a delicate balance between spin interactions and magnetic anisotropies, and can be tuned between the magnetic soliton lattice and uniform ferroelectric state by the application of magnetic and/or electric fields. This result is important for electric control of magnetism in a broad class of multiferroic materials with several coexistent magnetic orders.

ACKNOWLEDGMENTS

We acknowledge stimulating discussions with Dr. Tom Fennell. We acknowledge financial support from the Swiss National Science Foundation (Project No. 200020-169393) and from the German Research Foundation (DFG) through Project C03 of the Collaborative Research Center SFB 1143 (Project No. 247310070), and the Würzburg-Dresden Cluster of Excellence on Complexity and Topology in Quantum

TABLE I. The set of B_l^m parameters in units (meV) calculated using PCM.

B_l^m (meV)	
$B_2^0 = 4.30 \times 10^{-1}$	$B_6^0 = -6.24 \times 10^{-7}$
$B_2^2 = 2.63 \times 10^{-1}$	$B_6^2 = 2.26 \times 10^{-6}$
$B_2^{-2} = -5.53 \times 10^{-1}$	$B_6^{-2} = 2.68 \times 10^{-6}$
$B_4^0 = 4.79 \times 10^{-5}$	$B_6^4 = -1.53 \times 10^{-5}$
$B_4^2 = 5.64 \times 10^{-4}$	$B_6^{-4} = -6.77 \times 10^{-7}$
$B_4^{-2} = -1.43 \times 10^{-3}$	$B_6^6 = 2.15 \times 10^{-6}$
$B_4^4 = 4.61 \times 10^{-4}$	$B_6^{-6} = 6.21 \times 10^{-7}$
$B_4^{-4} = 4.01 \times 10^{-3}$	

Materials—*ct.qmat* (EXC 2147, Project No. 390858490). This work is based on experiments performed at the Swiss spallation neutron source SINQ, Paul Scherrer Institute, Villigen, Switzerland.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

APPENDIX A: POINT CHARGE MODEL CALCULATIONS OF CRYSTAL ELECTRIC FIELD

The CEF Hamiltonian of Dy^{3+} ions is given by

$$\mathcal{H}_{\text{CEF}} = B_l^m O_l^m, \quad (\text{A1})$$

where B_l^m are parameters of the Hamiltonian and O_l^m are Stevens operators. Due to low site symmetry of Dy^{3+} ions (C_s) in DyFeO_3 , the Hamiltonian includes 15 independent parameters. We performed point charge model calculations using the MCFE software [29]. The calculated B_l^m parameters are summarized in Table I. We report the B_l^m parameters of the CEF Hamiltonian using in the coordinate system with x, y, z parallel to the a, b , and c axes of the crystal [left panel in Fig. 1(e)], while the wave functions are given in the rotated coordinate system where the z axis points along the Ising axis of Dy moments [right panel, Fig. 1(e)]. This coordinate system can be obtained from the original one [$\mathbf{a} \parallel \mathbf{x}$, $\mathbf{b} \parallel \mathbf{y}$, and $\mathbf{c} \parallel \mathbf{z}$] by two consecutive rotations: (1) $\varphi = 61^\circ$ around $\mathbf{z}$ axis and (2) $\pi/2$ around the $\mathbf{y}$ axis.

To verify the accuracy of our PCM model, we calculated the saturation magnetization along the three crystal axes and found $M_a = 4.8 \mu_B$, $M_b = 8.8 \mu_B$, and $M_c < 0.3 \mu_B$. These results are in excellent quantitative agreement with the experimental data from Ref. [20] taking into account the simplicity of the model ($M_a = 4.4 \mu_B$, $M_b = 8.6 \mu_B$, and $M_c = 0.9 \mu_B$ at $T = 2$ K and $B = 7$ T). The difference in the c -axis signal likely arises from the weak FM component of the Fe subsystems, which was not included in the calculation. The wave functions of the ground state the and first excited doublets (which lies at 28.01 meV) are given by

$$|\psi_{\pm}\rangle_0 = 0.997|\pm 15/2\rangle + \dots, \quad (\text{A2})$$

$$|\psi_{\pm}\rangle_1 = 0.999|\pm 13/2\rangle + \dots. \quad (\text{A3})$$

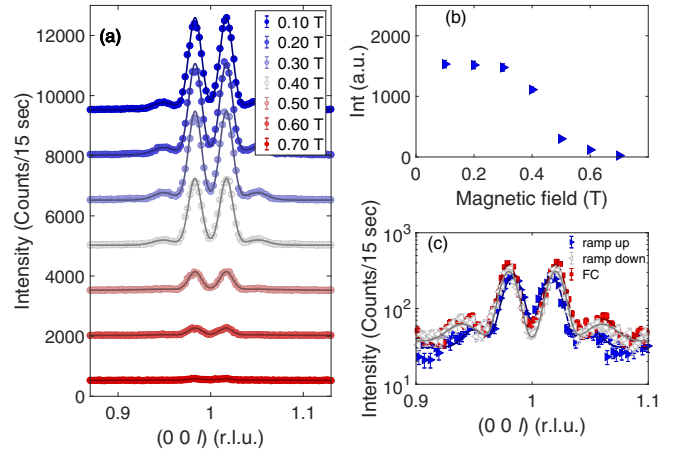


FIG. 6. (a) Neutron diffraction scans measured along $(0, 0, l)$ directions at $T = 2$ K and various fields applied along $[1, 1, 0]$ the direction as indicated in the legend. Data are stacked by 1500 units for clarity. (b) Intensity of $(0, 0, 1 \pm \delta)$ peaks extracted from the data in panel (a). (c) Scans along $(0, 0, l)$ direction taken at $T = 2$ K and $B = 0.6$ T state, prepared using different protocols. Blue curve was collected after ZFC and then sweeping the field to 0.6 T. Gray curve was collected after FC at 0.6 T. Red curve was measured after sweeping the field down to 0.6 T from 1.6 T.

The wave functions consist of almost pure $|\pm 15/2\rangle$ and $|\pm 13/2\rangle$ states, respectively, having only negligible admixture of other states. Thus, we conclude that Dy magnetism at low temperatures, < 10 K, can be well described by the pseudo- $S = 1/2$ model with an extreme Ising-like anisotropy.

APPENDIX B: NEUTRON DIFFRACTION IN $[110]$ FIELDS

Additional measurements were performed with the magnetic field applied along the $[110]$ direction. Figure 6(a) shows the scans taken at $T = 2$ K and various fields, $B \parallel [110]$ applied using the horizontal MA-7 cryomagnet. One can see that the behavior changes drastically as compared to the $B \parallel [001]$ case. The magnetic field rapidly suppresses the magnetic satellites, which become undetectable already at 0.7 T [Fig. 6(b)]; however, the propagation wave vector remains unchanged. To see whether the magnetic prehistory influences the observed signal, we performed scans at 0.6 T and 2 K using different cooling procedures: (1) ZFC and then apply 0.6 K field; (2) FC at 0.6 T; and (3) FC in 1.6 T and then decrease the field to 0.6 T. The results are summarized in Fig. 6(c). Our data show a small change in intensity of the satellites indicating a hysteretic behavior in agreement with Ref. [19]. Moreover, the propagation wave vector also exhibits a moderate change from 0.0165(2) (ZFC) to 0.0201(2) (FC).

APPENDIX C: EMPIRICAL FITS OF THE DIFFRACTION DATA

In Figs. 1(f) and 1(g) of the main text, we show temperature dependencies of the intensity and position of the primary AFM peaks, $(0, 0, 1 \pm \delta)$. They were obtained by empirical fits to the data using a sum of Gaussian functions peaked at odd harmonics, $n = 1, 3, 5$, and 7. The broad tail at $l > 1.1$

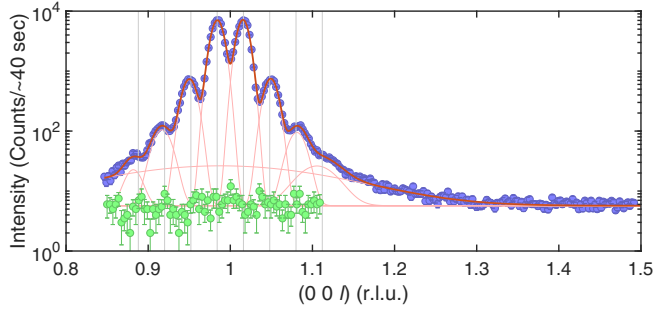


FIG. 7. Neutron diffraction scans measured along the $(0, 0, l)$ directions at $T = 1.7$ and 4.5 K. The data were corrected for the absorption. The data are fitted as a sum of nine Gaussian functions describing first, third, fifth, and seventh harmonics and a broad peak responsible for the long tail at $l > 1.1$. Blue and green dots show data taken at 1.7 and 4.5 K, the dark red line is the total fit and the light red curves demonstrate the decomposition of the total fit to individual Gaussians. The incommensurability parameter is best determined for the first two peaks at $(0, 0, 1 \pm \delta)$. Vertical gray lines show the calculated positions of the higher harmonics based on the δ value obtained for the first harmonics.

was empirically described by a broad Gaussian centered at $l = 1$. An example of such fit for the base temperature is shown in Fig. 7.

APPENDIX D: DETAILS OF MONTE CARLO SIMULATIONS

To simulate the observed diffraction profile, we considered a one-dimensional Ising chain, the diffraction pattern of which was calculated with the help of the Monte Carlo method. The standard fast Fourier transform (FFT) was employed to calculate the diffraction pattern of the sampled chain. Therefore, it was convenient to set the chain size N as a power of two, 2^n , in order to take advantage of the FFT algorithm. In our case, $N = 2^{19} = 524\,288$ the number of spins was chosen and deemed sufficiently large. The calculated Fourier transform was then scaled using the magnetic atomic form factor of Dy^{3+} to better approximate the experimental neutron scattering intensity. Additionally, an absorption correction was applied to the calculated diffraction intensity, to account for strong absorption of Dy nuclei. The absorption was estimated using the DAVE software [30], assuming the cylindrical shape of the sample. Finally, to obtain the calculated intensity, the corrected Fourier transform was convoluted with the experimental resolution function taken as a Gaussian with $\sigma = 0.0073$ r.l.u. (calculated using takin [31] software).

We considered an AFM spin chain with domain wall-type defects in a manner of $\cdots \uparrow \downarrow \uparrow \downarrow \uparrow \downarrow \cdots$. In order to conveniently compare the results with DyFeO_3 , the unit cell of the chain was selected to include two spins. Because of that, in the absence of defects, the main reflection occurs at $L = 1$, corresponding to $\mathbf{Q} = (0, 0, 1)$ in DyFeO_3 . Once the periodic domain walls are introduced, the peak splits into IC satellites at $L = 1 \pm \delta$ along with corresponding higher-order harmonics, as discussed in the main text. The distance between the defects (i.e. the size of the AFM domains) directly controls the δ parameter.

APPENDIX E: THEORY OF THE IC STATE IN DyFeO_3

In this section, we provide additional details regarding the theory calculations presented in Sec. III D.

1. Weak coupling approximation

In the absence of applied fields, the direction of the Néel vector is close to the b axis. Expanding free energy in powers of $\varphi(z) = \frac{\pi}{2} - \phi(z)$, we obtain

$$\left[-J \frac{d^2}{dz^2} + K_x\right] \varphi(z) = 2g \sum_n Q_n \delta(z - z_n). \quad (\text{E1})$$

The solution is

$$\varphi(z) = \frac{g}{\sqrt{JK_x}} \sum_n Q_n e^{\frac{|z-z_n|}{l}}, \quad (\text{E2})$$

where $l = \sqrt{\frac{J}{K_x}}$.

Substituting $\varphi(z)$ into Eq. (2) of the main text, we obtain free energy of the domain wall array

$$F - F(\varphi = 0) = N f_{\text{Dy}}^{(0)} - \frac{g^2}{\sqrt{JK_x}} \sum_{n,m} Q_n Q_m e^{\frac{|z_n - z_m|}{l}}, \quad (\text{E3})$$

where $F(\varphi = 0)$ is the free energy of the uniform state with $n \parallel b$ and N is the number of domain walls. The terms with $n = m$ in the double sum over domain wall positions describe the self-interaction that lowers the domain wall free energy: $f_{\text{Dy}}^{(0)} \rightarrow f_{\text{Dy}} = f_{\text{Dy}}^{(0)}(1 - \lambda)$, $\lambda = \frac{g^2}{f_{\text{Dy}}^{(0)} \sqrt{JK_x}}$ being the dimensionless coupling constant. The interaction between two domain walls (per unit area of the wall) is

$$U(z_n - z_m) = -\frac{2g^2}{\sqrt{JK_x}} Q_n Q_m e^{\frac{|z_n - z_m|}{l}}. \quad (\text{E4})$$

This result can also be obtained by integrating “the screened Coulomb potential,” $-\frac{g^2}{\pi J} Q_n Q_m \frac{1}{r} e^{-r/l}$, over the domain wall surface. The “screening” originates from the gap in the magnon spectrum. This shows that Dy domain walls behave like particles that interact by emitting and absorbing virtual magnons propagating through the Fe magnetic sublattice.

The free energy of an equidistant domain wall array with $z_n = nd$ is given by

$$F - F(\varphi = 0) = N f_{\text{Dy}}^{(0)} \left(1 - \lambda \tanh \frac{d}{2l}\right), \quad (\text{E5})$$

and the distance d between the domain walls is found by minimizing the free energy of the domain wall array per unit length.

2. Coupling to electromagnetic field

The interaction with the electric field, E_0 , and magnetic field, H_0 , applied along the c axis is described by

$$f_{\text{int}} = -E_0 P_z - H_0 M_z + \frac{2\pi}{\epsilon} (P_z - \bar{P}_z)^2 + 2\pi M_z^2, \quad (\text{E6})$$

where the electric polarization, P_z , and magnetization, M_z , are given by Eq. (3) in the main text. The last terms are, respectively, the depolarization and demagnetization energies (here, we use the CGS system of units). We assumed, for simplicity,

that the sample has the shape of a thin film of thickness L_z in the c direction and that the electric field is applied by ideal electrodes attached to its upper and lower surfaces normal to the c axis. The electric field, E_z , in Eq. (3) is related to the applied electric field, $E_0 = \frac{U}{L_z}$, U being the applied voltage, by

$$E_z = E_0 + \frac{4\pi}{\epsilon}(\bar{P}_z - P_z), \quad (\text{E7})$$

where $\bar{P}_z$ is the average polarization and $\epsilon = 23$ is the background electric permittivity [32], which follows from the fact that the z component of the displacement field, $D_z = \epsilon E_z + 4\pi P_z$, is constant and equal to its average value, and from $\bar{E}_z = E_0$. In the uniform state with $\bar{P}_z = P_z$ the depolarization energy is zero, because the surface charges are screened by electrodes. On the other hand, the depolarization energy is nonzero in the domain wall array state due to the electric charges induced in the bulk. The close competition between the IC and ferroelectric states, and the large depolarization field value, $E_{\text{dep}} = \frac{4\pi P_0}{\epsilon} \approx 73 \text{ kV cm}^{-1}$, make the depolarization energy important.

The demagnetization field induced by the weak ferromagnetic moment, $4\pi M_{\text{WFM}} \approx 270 \text{ G}$, as well as the magnetic moment induced by applied electric fields, are small, so that the demagnetization effects are only important for very weak magnetic fields and can be neglected. There is also a term quadratic in the applied magnetic field, $\frac{\chi_{\perp}}{2}[(\mathbf{n} \cdot \mathbf{H})^2 - H^2]$, where $\chi_{\perp}$ is the transverse magnetic susceptibility of the antiferromagnetically ordered state of Fe spins. However, for the magnetic fields used in our experiment, this term is relatively small. Furthermore, for $H \parallel c$, $\mathbf{n}$ rotates in the ab plane and is perpendicular to $\mathbf{H}$. The interaction of the Ising-like Dy spins parallel to the ab plane with $H \parallel c$ can be neglected.

3. Numerical simulations of the IC states

The calculation of the IC state spin configuration in the presence of applied electric and magnetic fields is complicated by the fact that the depolarization energy depends on the average polarization, $\bar{P}_z$, which itself depends on the applied fields, $\phi(z)$ and domain wall coordinates. This problem does not arise if only E_0 or H_0 is applied, as in these cases $\bar{P}_z = 0$. In the following, we describe the self-consistent calculation of $\phi(z)$ and domain wall positions in applied electromagnetic fields.

Minimizing the sum of Eqs. (2) and (E6), we obtain the following equation for $\phi(z)$ in the IC state:

$$\begin{aligned} & -J \frac{d^2 \phi}{dz^2} - \left(K_x + \frac{4\pi P_0^2}{\epsilon} \right) \cos \phi \sin \phi - K'_x \cos^3 \phi \sin \phi \\ & + P_0 \left(E_0 + \frac{4\pi \bar{P}_z}{\epsilon} + \frac{4\pi \alpha}{\epsilon} H_0 \bar{\sigma} \right) \sigma \sin \phi \\ & + H_0 \left(M_{\text{WFM}} - \frac{8\pi \alpha P_0}{\epsilon} \right) \sin \phi \\ & + 2g \sum_n Q_n \sin \phi(z_n) \delta(z - z_n) = 0, \end{aligned} \quad (\text{E8})$$

where $\bar{\sigma}$ is the average value of the Dy order parameter. Here, we took into account that the average polarization, $\bar{P}_z$, is a

functional of $\phi(z)$. For numerical simulations, it is more convenient to do minimization at constant $\bar{P}_z$. The corresponding functional that leads to the same equation for $\phi(z)$ has the form,

$$\begin{aligned} F' = \int dz & \left[\frac{J}{2} \left(\frac{d\phi}{dz} \right)^2 + \frac{1}{2} \left(K_x + \frac{4\pi P_0^2}{\epsilon} \right) \cos^2 \phi + \frac{K'_x}{4} \cos^4 \phi \right. \\ & - \frac{P_0}{\epsilon} (\epsilon E_0 + 4\pi \bar{P}_z + 4\pi \alpha H_0 \bar{\sigma}) \sigma \cos \phi \\ & - H_0 \left(M_{\text{WFM}} - \frac{8\pi \alpha P_0}{\epsilon} \right) \cos \phi \\ & \left. + \sum_n \left(f_{\text{Dy}}^{(0)} - 2g \sum_n Q_n \cos \phi \delta(z - z_n) \right) \right]. \quad (\text{E9}) \end{aligned}$$

The depolarization energy modifies the second-order magnetic anisotropy and the weak ferromagnetic moment. Importantly, F' depends on E_0 and $\bar{P}_z$ through the displacement field, $D_z = \epsilon E_0 + 4\pi \bar{P}_z$. First, we find $\phi(z)$ for a given D_z and positions of the domain walls, calculate $\bar{P}_z$ and $\bar{\sigma}$, minimize the free energy density with respect to the domain wall positions and replot it as a function of E_0 .

4. Parameter choice

Simulations are performed, for $J = 4.5S^2 \text{ meV}$, $S = 5/2$ being the spin of the Fe ion, similar to that found for YFeO_3 and other orthoferrites [12,33,34]. We chose $f_{\text{Dy}}^{(0)} = 2.6 \text{ K}$ (the ordering temperature of Dy spins, $T_{\text{Dy}} \sim 4.5 \text{ K}$). The choice of the Fe-Dy coupling constant g and the anisotropy parameter K_x was made as follows. We found that for $\lambda \gg 1$, the IC state period strongly increases with an applied magnetic field $H \parallel c$

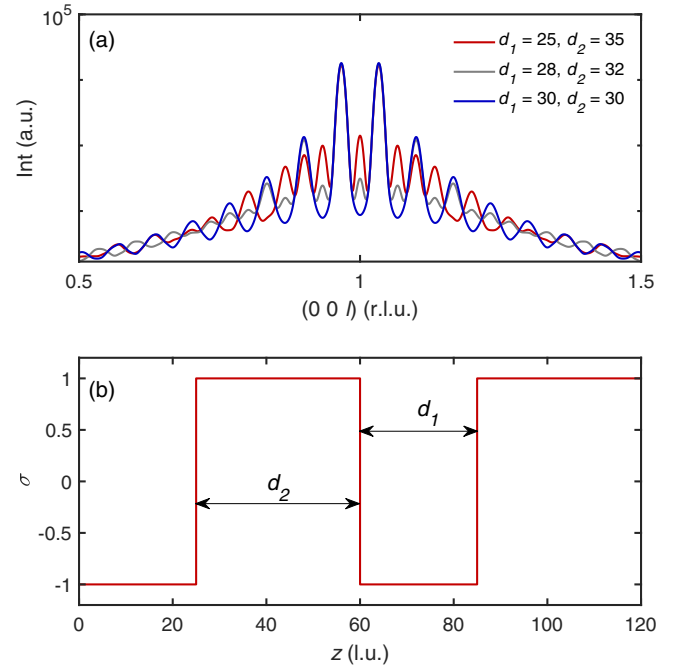


FIG. 8. Temperature dependence of integrated magnetic intensity measured at different fields applied along c axis.

before this state disappears at ~ 2 T, which is not observed in our experiment. On the other hand, when λ is close to the critical value 1, the IC state disappears at very low applied fields. We use $g = 8.5$ K and $K_x = 0.5$ K, corresponding to $\lambda = 2.2$, for which the period changes between $58c$ in zero field and $62c$ at $H_{c1} = 2.2$ T, when the IC state starts to disappear in our experiment. In calculations, it undergoes a first-order transition into a uniform ferroelectric state with $\phi > 0$ at H_{c1} . Tokunaga *et al.* [10] found that spontaneous polarization saturates and the magnetic field dependence of P solely results from the linear magnetoelectric effect above $H_{c2} \approx 2.6$ T, corresponding to the disappearance of IC peaks in our experiment, which suggests that at H_{c2} the Néel vector becomes parallel to the a axis. We reproduce two first-order transitions at H_{c1} and H_{c2} by adding the fourth-order anisotropy term with $K'_x = -0.32$ K. The slope of $P(H)$ above H_{c2} is reproduced using $\alpha = 0.0072$ in dimensionless CGS units [$\alpha = 0.024$ reported in Ref. [10] is apparently given in $\frac{\mu\text{C}}{\text{cm}^2\text{T}}$, which can be deduced from Fig. 3(b) in that paper]. For $\lambda = 2.2$, the weak

coupling approximation and numerical simulations give close results.

APPENDIX F: DOMAIN WALL ARRAY DIMERIZATION

In Sec. III D, we demonstrate for $H_0 \neq 0$, $E_0 = 0$, as well as for $H_0 = 0$, $E_0 \neq 0$, the domain wall array remains equidistant and has a zero net polarization [Fig. 5(d)]. However, when both fields are nonzero, our theory predicts that the domain wall array become dimerized [Fig. 5(g)]. This behavior should strongly affect observed diffraction signal. To illustrate this, we performed simple calculations assuming different degrees of dimerization as shown in Fig. 8. For $d_1 = d_2$, the calculated curve consists of multiple peaks centered at odd harmonics of the primary propagation wave vector δ . In contrast, when finite dimerization is present, one can clearly see appearance of even-order harmonics along with the commensurate peak at (001). The intensity of the new peaks is controlled by the strength of dimerization.

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