Electric field-induced domain wall motion in spin spiral multiferroics

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Switching in magnetic materials gives rise to rich physical phenomena and lies at the heart of their technological applications. Although domain wall motion in ferro- and antiferromagnets has been studied \cite{1-3}, in spiral magnets it is still poorly understood despite 20 years of active research since the discovery of spiral multiferroics \cite{4-8}. The problem of the domain wall motion in a spiral magnet is a compelling one, the more so the magnetic domain walls in cycloidal spiral phase are also ferroelectric, thus enabling electric control of magnetism, i.e. domain wall motion under the action of an external electric field. Phase transition to a spiral phase leads to a formation of chiral domains with opposite spin rotation senses, that are separated by chiral domain walls. Spiral order breaks inversion symmetry and induces a ferroelectric polarization, whose sign is determined by the chirality of the domain. Thus the spiral order allows for the manipulation of spins via an external electric field. Here we study domain wall motion in magnets with spiral ground state, that are the most basic non-collinear magnets. We formulate a simplified variational model and derive the equation of motion for the domain wall driven by an external electric field. The results are corroborated with atomistic spin dynamics simulations. The results suggest a linear dependence of the wall speed on the external electric field, and a peculiar dependence on the system geometry and domain structure.

\textbf{Introduction} — Magnetic frustration, or competition between various spin interactions, plays a fundamental role in magneto- and multiferroic properties \cite{9}. Geometric frustration or competing nearest and next-nearest-neighbor exchange interactions can result in peculiar spin ordering, skyrmions and other non-collinear spin textures, and could lead to polar lattice distortions \cite{5,10-12}. This scenario is the ideal playground for complex spin textures and magneto- and electric coupling to appear \cite{5,13,14}. In particular, the study of the DW and skyrmion dynamics has attracted experimental and theoretical interest in the recent years \cite{15,16,18}.

In this Letter we study domain wall motion in the most basic non-collinear magnet, one with a cycloidal spiral spin ordering, stabilized by competing exchange interactions, as in RMnO\textsubscript{3}, YMn\textsubscript{2}O\textsubscript{5}, MnWO\textsubscript{4} or CuO \cite{4,5}. Our aim is to investigate the motion of a DW, which we refer to as a chiral DW \cite{19}. In this context “chiral” refers to a system that is not identical to its mirror image and, in the particular case of the spiral spin order, a chiral domain is a region where the rotation sense of the spins is well-defined. Therefore, chirality is strictly related to the spin rotation sense, and a chiral DW is the region of separation between two domains of opposite chirality, i.e. opposite spin rotation senses.

While the concept of chirality in motion of chiral DWs have recently attracted attention \cite{20-23}, to our knowledge, electric field driven switching in spiral magnets has not been studied. In a cycloidal spiral spin configuration, where the spin rotation plane contains the wave vector, inversion symmetry is broken, and a ferroelectric polarization emerges through inverse Dzyaloshinskii-Moriya (DM) effect \cite{23,27}.

We proceed in the following way: we propose an Ansatz for the chiral DW profile and, using a variational approach, obtain the equations of motion for the DW. We study the cases of one single DW and two DWs moving in an external electric field in a quasi-1D geometry. The results of the model are then compared with the atomistic spin dynamics simulations. The last part of the Letter is dedicated to the discussion of the simulations. We observe that, even when starting with a single-DW configuration, another domain is nucleated at the boundary of the sample in the presence of the electric field. DWs do not behave as isolated objects and move according to the presence of other walls, as seen from the comparison of the one-DW and two-DW cases. The results suggest that the domains of polarization opposite to the external field shrink and finally disappear with the emission of magnons. The domain wall velocity in a linear response regime is proportional to the driving electric field and inversely proportional to the Gilbert damping and the distance to the boundary. No Walker breakdown is observed in simulations.

\textbf{The Model} — In order to study the motion of the chiral DWs, we consider a quasi-1D model, where a magnetization M(x) continuously varies along x through an interval of total length L. M can be directed along all the three spatial directions.

The dynamics of the spiral magnet is described by the Lagrangian density \(L\) \cite{13,28},

\begin{equation}
L = \alpha A(M) \cdot \dot{M} - J(\nabla M)^2 - J'(\nabla^2 M)^2 + \gamma [E \times \mathbf{x}] \cdot [M \times \nabla M] + k_z M_z^2,
\end{equation}

where \(\alpha\) is a constant that has the dimensions of an action, the first term describes the precession of the magnetization, with \(A\) acting as a vector potential so that \(\nabla_M \times A(M) = M\) \cite{28}. \(J < 0\) and \(J' > 0\) describe competing magnetic exchange constants, and produce spatial variation of the magnetization with the wavevector \(k_z\).
where \( \phi \) that captures the essential physics of the DW motion: we assume the following Ansatz for the angles with a single DW at the position \( \bar{x} \) that describes the energy dissipation. The term \( \delta \) kinetic energy proportional to the spin-orbit coupling constant. The spiral order and the field, resulting from DM interaction and proportional to the spin-orbit coupling constant. \( k_z \) is the hard axis anisotropy constant that forces spins into the \( xy \)-plane. As we will see from the following, the main contributions come from the domains, which suggests the dynamics to be largely independent of the DW type.

We can express the magnetization \( M \) in spherical coordinates, in terms of the angles \( \theta(x) \) and \( \phi(x) \) (just \( \theta \) and \( \phi \) in the following, for simplicity)

\[
\mathcal{L} = \alpha \dot{\phi} \cos \theta - J(\dot{\phi} \cos \theta + \phi \dot{\phi} \sin \theta) + J' \dot{\phi} \sin \theta + \gamma E \dot{\phi} \sin^2 \theta + k_z \cos^2 \theta,
\]  
(2)

This is a continuous generalization of \( J_1 - J_2 \) model with competing nearest and next-nearest neighbor interactions. The term \( \dot{\phi} \sin^2 \theta \) is the only term from the expansion of \( \nabla^2 \mathbf{M} \) that will contribute to our model, as we will see in the following. We also introduce the Rayleigh dissipation functional density

\[
\mathcal{R} = \beta \mathbf{M}^2 = \beta [\dot{\phi}^2 \sin^2 \theta + \dot{\theta}^2]
\]  
(3)

that describes the energy dissipation. \( \beta \) is the damping constant and has dimensions of a momentum.

In order to describe a cycloidal spiral spin configuration with a single DW at the position \( \bar{x} \), as seen in Fig. 1, we assume the following Ansatz for the angles \( \phi \) and \( \theta \), that captures the essential physics of the DW motion:

\[
\phi = \begin{cases} 
\phi_0 - mx & \text{if } x < \bar{x} \\
\phi_0 - m\bar{x} + m(x - \bar{x}) & \text{if } x \geq \bar{x}
\end{cases}
\]

\[
\cos \theta = \begin{cases} 
\delta_1 & \text{if } x < \bar{x} \\
\delta_2 & \text{if } x \geq \bar{x}
\end{cases}
\]  
(4)

where \( m \) is the wave vector of the spiral, i.e. the angle between two nearby spins; \( \phi_0 \) is the initial phase for the angle and \( \delta_{1,2} \) (with \( \delta_{1,2} \ll 1 \)) represents the out-of-plane components of the spins. Here the shape of \( \phi \) is inspired by the usual spiral solution in the \( J_1 - J_2 \) model, however with a DW in the center.

The presence of the \( M_z \) components of the magnetization encoded by \( \delta_{1,2} \) is needed for the magnetization to rotate. Indeed, for the DW to move, spins must precess around \( z \) axis. Magnetization precesses around the effective field \( h_n = -\frac{\partial \mathcal{L}}{\partial \mathbf{M}} \) that for spins lying in \( xy \)-plane has no \( z \) component. The precession around \( z \) axis becomes possible because the magnetization acquires \( M_z \) component due to DM field, that has a component in \( xy \) plane perpendicular to the magnetization.

In the spirit of a variational approach, we integrate \( \mathcal{L} \) and \( \mathcal{R} \) over the sample. We continue to refer to the integrated Lagrangian and Rayleigh functions with \( \mathcal{L} \) and \( \mathcal{R} \) for simplicity.

\[
\mathcal{L} = \alpha \{ [\phi_0 \delta_1 x - (\phi_0 - 2m\bar{x}) \delta_2 (L - x)] - J m^2 [(1 - \delta_1^2) x + (1 - \delta_2^2) (L - x)] + m \gamma E \{ (1 - \delta_1^2) x + (1 - \delta_2^2) (L - x) \} + k_z (\delta_1^2 x + \delta_2^2 (L - x)) \}
\]  
(5)

\[
\mathcal{R} = \beta \left[ (\phi_0^2 + 4m^2 \bar{x}^2 - 4m \bar{x} \phi_0)(1 - \delta_2^2) (L - x) + \phi_0^2 (1 - \delta_1^2) x + \frac{\delta_1^2}{1 - \delta_1^2} x + \frac{\delta_2^2}{1 - \delta_2^2} (L - x) \right].
\]  
(6)

Results – We derive from the Lagrangian (5) and the Rayleigh functional (3) the equations of motion for the DW

\[
\frac{\partial \mathcal{L}}{\partial \xi} - \frac{\partial}{\partial t} \frac{\partial \mathcal{L}}{\partial \dot{\xi}} = \frac{\partial \mathcal{R}}{\partial \xi}
\]  
(7)

where \( \xi = \bar{x}, \phi_0, \phi_1, \phi_2 \). We first consider the equations for \( \delta_1 \) and \( \delta_2 \),

\[
\alpha \dot{\phi}_0 + 2 \Gamma_1 \delta_1 = \frac{2 \beta \delta_1}{1 - \delta_1^2},
\]

\[
\alpha (\dot{\phi}_0 - 2m \dot{\bar{x}}) + 2 \Gamma_2 \delta_2 = \frac{2 \beta \delta_2}{1 - \delta_2^2},
\]  
(8)

with \( \Gamma_{1,2} = (J m^2 \pm m \gamma E + k_z) \), however, as the electric field \( E \) is small with respect to \( J \), we can write \( \Gamma_1 \approx \Gamma_2 \approx \Gamma \). The shape of these equations suggest a solution composed of a transient part with a timescale of \( \beta/\Gamma \) and a steady state term. The small \( \delta^2 \) term in the denominator is neglected and the solutions for \( \delta_{1,2} \) are found as

\[
\delta_1(t) = A e^{\delta_1 t} - \alpha \frac{\phi_0}{2 \Gamma},
\]  

\[
\delta_2(t) = B e^{\delta_2 t} - \alpha \frac{\phi_0 - 2m \bar{x}}{2 \Gamma}.
\]  
(9)
When approaching the boundary. This also explains why the DW velocity diverges of the domain, so that spins in the larger domain rotate \( \delta \). We note that both \( \dot{\phi}_0 \) and \( \dot{x} \) are linear in the field. As we mentioned before, in the equation of motion we neglected terms except for the lowest order in \( \dot{\phi}_0 \) and \( \dot{x} \). It is now evident that the terms we retained are of the lowest order in the external field \( E \). Moreover, substituting Eq. (11), (12) into Eq. (9), we obtain

\[
\beta (4m \dot{x} - 2 \dot{\phi}_0) (L - x) + \gamma E = 0 \\
(2 \dot{\phi}_0 - 4m \dot{x}) (L - \bar{x}) + 2 \dot{\phi}_0 \bar{x} = 0. \tag{10}
\]

From these equations we obtain

\[
\dot{\phi}_0 = -\frac{\gamma E}{2 \beta \bar{x}}, \tag{11}
\]

\[
\dot{x} = -\frac{\gamma E}{4 \beta m \bar{x} (L - \bar{x})}. \tag{12}
\]

It is remarkable that the DW velocity is inversely proportional to the distance from the boundaries, \( x \) and \( L - x \). This can be understood from the fact that fewer spins need to rotate to move the DW when it is near the boundary. We note that both \( \dot{\phi}_0 \) and \( \dot{x} \) are linear in the field. According to Eq. (8), the rate of spin precession \( \dot{\phi}_0 \) in the first domain in the steady state (\( \delta_{1,2} = 0 \)) is determined by \( \delta_1 \). Analogously, in the second domain it is proportional to \( \delta_2 \). Thus the torque on spins due to \( M_x \) component is distributed according to the size of the domain, so that spins in the larger domain rotate less. This also explains why the DW velocity diverges when approaching the boundary.

The integration of Eq. (12) gives a cubic equation for \( x(t) \). In the limit of \( \bar{x} \ll L \), when the DW near the sample boundary, the \( x^3 \) term dominates and we obtain

\[
x(t) = \sqrt{x^2(t_0) - \frac{\gamma E}{2 \beta m} (t - t_0)} \tag{14}
\]

From now on, we will refer to \( \bar{x} \) as \( x \) for simplicity.

Two DWs — In real materials many DWs are present at the same time, so that when an electric field \( E \) is applied on the whole sample, all the walls move. In a quasi-1D picture, we can think of the pairs of DWs delimiting domains of polarization opposite to the applied electric field. We expect for DWs in such pairs to move towards each other under the action of the field, so as to drive the compression of ferroelectric domains of unfavorable polarization. In analogy with the previous case, we use a modified Ansatz,

\[
\phi = \begin{cases} 
\phi_0 - m x & x < x_1 \\
\phi_0 - m (x - x_1) & x_1 \leq x \leq x_2 \\
\phi_0 + m (x_2 - 2x_1) - m(x - x_2) & x > x_2 
\end{cases} \\
\cos \theta = \begin{cases} 
\delta_1 & x < x_1 \\
\delta_2 & x_1 \leq x \leq x_2 \\
\delta_3 & x > x_2 
\end{cases} . \tag{15}
\]

The notation is analogous to the single DW case, except for \( x_1 \) and \( x_2 \) that are the positions of the two DWs. We derive the equations of motion from the general Lagrangian [5] and Rayleigh functional [6], and truncate them to the lowest order in the external field \( E \). We obtain

\[
\dot{\phi}_0 = -\frac{\gamma E}{2 \beta x_1}, \ \dot{x}_1 = -\frac{\gamma E}{4 \beta m x_1}, \ \dot{x}_2 = \frac{\gamma E}{4 \beta m (L - x_2)} . \tag{16}
\]

We observe that, in the large-\( L \) limit, the expressions for \( \dot{\phi}_0 \) and \( x_1 \) become identical to those in the single DW case, Eq. (11), (12). It is interesting to notice that the speeds of the two walls are opposite in sign but generally not equal in modulus. We can understand the dependence of the speed of the walls on their position by focusing on the spin texture. When a DW moves, the sizes of the two domains that are separated by the wall change. In the growing domain, all the spins must be rotated by the same angle every time the DW moves by a unit cell, this costs energy. With the size of the domain the number of spins that must be rotated increases, and the torque, produced through the action of the electric field, is distributed on all spins. Thus the spins in the larger domains rotate less, and the corresponding walls move slower, i.e., the speed of the walls depends on their position.

By integrating the equations for the two walls we obtain

\[
x_1(t) = \sqrt{x_1^2(t_0) - \frac{\gamma E}{2 \beta m} (t - t_0)} \\
x_2(t) = \sqrt{(L - x_2(t_0))^2 - \frac{\gamma E}{2 \beta m} (t - t_0)} . \tag{17}
\]

Simulations — In order to validate our model, we performed atomistic spin dynamics simulations using the UppASD code [30] (see Methods section for details).

As we discussed before, in a quasi-1D picture and in the presence of an electric field, the energy of a domain is linear in its size, which could be seen as a linear potential between the walls. DWs associated with domains of the polarization opposite to the external field move in pairs. This is observed in our simulation where a new
FIG. 2. (a) Temporal evolution of a polarization profile \((\pm P_y\text{ in red, } -P_y\text{ in violet, time indicated in ps}).\) A chiral DW is initially placed at the center (marked with a dashed black line), and another one is nucleated at the right boundary. (b) Profiles of \(\phi(r)\), the polar angle characterizing the magnetization, plotted in uniform timesteps. Colorscale encodes different times. At \(t=0\) ps (purple), the angle profile resembles the proposed Ansatz for the single-DW case. The linear behaviour \(\phi(r) = mr\) corresponds to a spiral domain, and the V-shaped cusps correspond to chiral (and ferroelectric) DWs.

DW is formed at the right boundary. The DW at the center and at the boundary both move, however the wall in the center is slower, as seen in Fig. 2(a). This is due to the greater number of spins that have to be rotated by the torque arising due to the action of the \(E\) field for the central wall to be moved. The two walls move towards each other, collide and annihilate at approximately 70 ps, leaving the system in a single domain state. Upon collision, magnons are excited and propagate away from the collision site, as seen in the Supplementary GIF. Magnons have oscillating signatures in the polarization profile, that propagate away from the collision site. The magnon emission may slightly alter the trajectory of the walls and thus generate a small difference with our model.

The rotation of the spins can be visualized by plotting \(\phi(r)\), cf. Fig. 2. Straight lines \(\phi(r) = \pm mr\) represent uniform spin spiral and correspond to chiral domains, while V-shaped cusps are the DWs. Just after \(t=0\), a V-shape appears at the right boundary, indicating a nucleation of a second domain with \(P\) along the external field. The large distances between the right cusps at subsequent steps indicate large initial speed of the right DW, that is consistent with the discussion following Eq. (13) and the fact that the right domain is very narrow. As the nucleated domain on the right grows, the wall gets slower and the distances between the cusps decrease. The wall initially at the center starts with a lower velocity and just slightly accelerates while moving to the right.

We now compare the motion of the two walls with the predictions of Eq. (17). The simulated snapshots of the polarization texture at different times are shown in Fig. 2(a). Fig. 4 reports the DW trajectories from the simulations (dots) and the behaviour predicted by the model (solid lines).

We find good agreement between the simulation and the theory. Differences near \(t=0\) may be due to a transient effect that is beyond our steady state solution. At large \(E\) fields the differences between our linear-response theory and the simulations become more visible, as seen in the Fig. S1.

Fig. S2 shows the simulated time-dependent polarization that results from DW motion in Fig. 2. The initial polarization is zero as initially the two DWs with opposite chiralities and hence opposite polarizations have the same size. When the walls start moving, the domain with polarization along the field grows, and the polarization decreases towards negative values until it saturates when only one domain is left. Our model is also able to predict qualitatively the polarization evolution Fig. S2 however it does not reproduce some of the details. As the DWs move the overall contribution to the polarization changes accordingly (see Fig. 3 for reference) and it will be

\[
\mathbf{P}(t) \propto x_1(t) - x_2(t).
\]  

Eq. (18) shows how the \(t\)-dependent polarization is related to the position of the walls.

Polarization evolution obtained in simulations, Fig. S2 matches well with Eq. (18).

Conclusions – In this Letter, we discussed the dynamics of chiral domain walls driven by an external electric field. By using a variational approach, we obtained the equations of motion. We found that the speed of the wall is proportional to the external electric field, and has a peculiar dependence on the domain structure and the geometry of the system. The out of plane components of the magnetization are induced via DM interactions and drive the dynamics. The dependence on the domain structure is rationalized in terms of the torque distributed...
between spins in different domains, thus spins in smaller domains rotate faster. Contrary to Walker breakdown in ferromagnets [1], here we find a monotonic increase of the DW velocity with the driving field.

Our atomistic spin dynamics simulations validate the model and provide indications beyond linear response regime. The agreement between the model and the simulation corroborates our results, confirming the dynamics we predicted for the case of two domain walls moving towards each other.

The dynamic response of a chiral domain wall to an external electric field has, to our knowledge, never been modelled before. We hope the work will inspire future experimental research in spiral multiferroics potentially leading to new applications and technology. Evolution of truly 3D domain structures and magnets beyond currently considered easy plane case would be interesting to study in the future. Possible boundary effects on domain nucleation and effects due to proximity of two walls may also be interesting.

FIG. 4. Trajectories of DWs under applied electric field. Comparison between the analytical model (solid lines) and simulation (dots) of the motion of the two domain walls at positions $x_1$ and $x_2$.

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SUPPLEMENTARY INFORMATION – Methods

Modelling

Simulations are performed on 2D a square lattice with spin 1 magnetic ions with nearest neighbour interaction of $|J| = 0.034641$ mRy $\approx 0.47$ meV and next-to-nearest neighbour interaction of $|J'| = 0.01$ mRy $\approx 0.14$ meV. Dzyaloshinskij-Moriya vectors are reported in Table S1. A supercell of size $84 \times 24 \times 1$ unit cells, with periodic boundary conditions along $y$ and open boundary conditions along $x$ and $z$, has been used. Simulations have been performed at $T = 0$ K and 0.005 damping. 100 ps of evolution of the spin system have been simulated with 10000 steps, saving the texture every 100 iterations. We mimick the action of the external electric field through the DM interaction,

$$H_{DM} = \alpha [r_{12} \times P] \cdot [S_1 \times S_2],$$

where $r_{12}$ is a vector connecting sites 1 and 2, $P$ is a ferroelectric polarization induced by the displacement of an oxygen linking magnetic ions 1 and 2. In a paramagnetic state the system is centrosymmetric and $P = 0$. The application of an external electric field leads to $P = \chi E$, that produces the DM-like term in the Hamiltonian, $H_{DM} = D \cdot [S_1 \times S_2]$ with $D = \alpha \chi [r_{12} \times E]$. It is this value of $D$ that we use in UppASD to simulate the external electric field.

**Atomistic spin dynamics simulations**

Simulations are done in a quasi-2D geometry with classical and with the Hamiltonian consistent with Eq. (5) and with a magnetic spiral ground state. We started with a state with two domains of opposite chiralities separated by the DW at the center, as seen in $t = 0$ panel of Fig. 2. While applying an electric field (see Methods Section for details), we simulate the dynamics governed by the LLG equation [31],

$$\frac{dm_i}{dt} = -\frac{g}{1 + a^2} m_i \times (B_i + b_i(t)) - \frac{g}{m} a \frac{1}{1 + a^2} m_i \times (m_i \times (B_i + b_i(t)))$$

where $m_i$ is the magnetic moment at site $i$, with modulus $m$, the coefficient $g$ is the gyromagnetic ratio, $a$ is the damping parameter. $B_i$ is the magnetic field acting on the moment $m_i$, and $b_i(t)$ is a stochastic magnetic field that mimics the effect of finite temperature by applying random torques on spins.

The simulation parameters were $|D_{DM}| = 0.0008 \ldots 0.003$ meV, damping of 0.005 meV and $T=0.0$. As DM interaction is increased, the agreement with the linear-response model worsens, but qualitative agreement remains.

For the lowest values of DM constant, $D_{DM} = 0.005$ meV and $T = 0$ the DW at the right boundary does not appear. If the temperature is increased to $T = 0.5$ K, the second wall appears and moves similarly to the previous case. Hence thermal fluctuations help the wall nucleate. Zero damping at zero T does not make the second wall appear at this DM.

$(0 \ T) \ DM = 0.005$ meV and damping of 0.05 (10 times higher than before) give the same behaviour as the lower fields.

| $r_{ij}^x$ | $r_{ij}^y$ | $r_{ij}^z$ | $D_{ij}^x$ | $D_{ij}^y$ | $D_{ij}^z$ |
|----------|----------|----------|-----------|-----------|-----------|
| 0.5      | 0.5      | 0.0      | 0.0       | 0.0       | -0.001    |
| -0.5     | 0.5      | 0.0      | 0.0       | 0.0       | 0.001     |
| 0.5      | -0.5     | 0.0      | 0.0       | 0.0       | -0.001    |
| -0.5     | -0.5     | 0.0      | 0.0       | 0.0       | 0.001     |

TABLE S1. DM vectors for the square lattice
FIG. S1. Domain wall trajectories for different field strengths. When the strength of the $E$ field increases, the trajectories deviate from the model derived in the leading order in $E$. 
FIG. S2. Time dependence of the polarization. The dependence from the model ($\delta P \sim x_1 - x_2$) in green; the same using the DW coordinates from the simulation shown with red dots; the simulated polarization integrated over the sample in blue.