Multiferroic materials for spin-based logic devices

Rogerio de Sousa\textsuperscript{1} and Joel E. Moore\textsuperscript{1,2}

\textsuperscript{1}Department of Physics, University of California, Berkeley, CA 94720
\textsuperscript{2}Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720

(Dated: April 9, 2008)

Logical devices based on spin waves offer the potential to avoid dissipation mechanisms that limit devices based on either the charge or spin of mobile electrons. Multiferroic magnetoelectrics, which are materials that combine ferroelectric and magnetic order, allow direct switching of magnetic order and hence of spin-wave properties using an applied electric field. The intrinsic coupling between polarization and magnetic moments, generated by strong electronic correlations in these multiferroic materials, is argued to provide new approaches to spin-wave injection and spin-wave switching using applied voltages with no external magnetic field. These effects are shown to arise in a phenomenological Landau theory of coupled electronic and magnetic orders in multiferroic BiFeO\textsubscript{3}, and found to depend subtly on differences between the crystalline and film states of this material.

I. INTRODUCTION

Some limitations of current semiconductor logic devices, such as their relatively high power dissipation per operation, may be removed in devices that use electron spin rather than electron charge to carry out logic operations. While ordered domains of electron spin are currently the basic element of most information storage devices, information manipulation (i.e., logic operations) is currently accomplished using electron charge rather than spin. Spin-based logic is a relatively new field and, at the present time, has not convincingly demonstrated advantages in practice over ordinary charge-based logic. In many cases, it seems that devices using spin currents carried by itinerant electrons suffer from the same dissipation mechanisms as ordinary electronic devices.

However, ferromagnetic and antiferromagnetic insulators provide another route to spin currents: they support “spin waves” that can transport spin and hence information over macroscopic distances, without accompanying transport of charge. This article discusses the relevant physics of “multiferroic magnetoelectric” materials that allow direct electrical control of the spin-wave propagation, at room temperature and without external magnetic fields. Recently considerable effort has gone into demonstrating basic logic operations with spin waves\textsuperscript{1,2}, as the dissipation mechanisms in this approach to logic and the interactions between spin waves are not well understood. Multiferroic materials with both magnetic and ferroelectric order address several of the most pressing problems in making such spin-wave devices practical for applications: in the near term, they allow electrical creation and detection of spin waves in a compact volume and can hence serve as efficient converters between electronic and spintronic logic. In the longer term, multiferroic materials may enable new designs for spintronic logic devices.

The value of multiferroic magnetoelectrics is that these strongly correlated materials couple strongly to applied electric fields, because of their ferroelectric character, while at the same time showing magnetic order that enables propagation of spin waves. Furthermore, the magnetic and ferroelectric orders are strongly coupled, as in the recent demonstration of room-temperature switching of magnetization direction by an electrical field in\textsuperscript{3}. As an example of how this enables spin-wave manipulation, we will show that the properties of a spin wave at wavevector $k$, such as its group velocity, depend on the angle between $k$ and the Néel magnetization direction. The result of this combination of effects is that electrical switching of the magnetization direction modifies the spin-wave propagation sufficiently strongly to stop or pass spin waves depending on the electrical state. This functionality is similar in concept to the function of ferroelectric materials as electrooptic modulators\textsuperscript{4}. While multiferroic materials have a relatively long history\textsuperscript{5,6,7}, recent advances in fabrication and control of these materials have led to a considerable resurgence of interest\textsuperscript{7}.

The theoretical calculations described in this paper are based primarily on a time-dependent Ginzburg-Landau model for the free energy of multiferroic materials. The dynamical equations combine the familiar Landau-Lifshitz dynamics of ferromagnetic and antiferromagnetic materials, the standard macroscopic free energy for polarization modes in ferroelectric materials, and additional terms that couple the two orders. The equations are solved to yield the spectra of coupled polarization and spin-wave modes. In choosing the microscopic details of the multiferroic couplings, we focus for simplicity on the material BiFeO\textsubscript{3}. In bulk crystals of BiFeO\textsubscript{3}, cycloidal antiferromagnetic order is established at 650 K, well below the ferroelectric transition at 1120 K. The next section introduces the basic model for these crystals and explains one key result for spin-wave manipulation in these crystals well below the transition temperature: a zero-wavevector electric perturbation, such as an AC electric field, couples to finite-wavevector spin waves, mediated by the cycloidal magnetic structure in the ground state. Films of BiFeO\textsubscript{3} differ significantly from bulk crystals: they show uniform antiferromagnetic order with a weak ferromagnetic component, rather than cycloidal order. Since the ability to create films is of considerable importance for applications, Section III gives an overview of how the model and results need to be modified for BiFeO\textsubscript{3}. 

Theoretical considerations.
films, and concentrates on how the switching process already demonstrated in one experiment will modify spin wave propagation. While our focus will be on BiFeO$_3$ because it is the predominant room-temperature multiferroic material, similar models can be applied to other low-temperature multiferroic materials as well.

II. MAGNETOELECTRIC COUPLING IN BULK CRYSTALLINE BIFEO$_3$: ELECTRONICALLY ACTIVE SPIN WAVES

The first step in the theoretical description of this material is to construct an appropriate phenomenological free energy. At room temperature, the system is far below its critical temperature for antiferromagnetic and ferroelectric transitions, so that thermal fluctuations can be ignored in the dynamics. The long-period incommensurate cycloid is produced in our model by a Lifshitz term in the effective Ginzburg-Landau free energy; the existence of such a term was previously argued as the simplest symmetry-allowed explanation for the cycloidal order that is observed in experiments unless a very large magnetic field is applied (around 18 Tesla). The model free energy is

\[ F = GL^4 + \frac{AL^2}{4} + \frac{c'}{2} \sum_i (\nabla L_i)^2 \]

\[ -\frac{a P \cdot [L(\nabla \cdot L) + L \times (\nabla \times L)] - P \cdot E}{2} \]

\[ + \frac{r M^2}{2} + \frac{a P^2}{2} + \frac{u P^2}{4} + \frac{a_{\perp} (P^2 + P_y^2)}{2}. \] (1)

Here \( L = |M_1 - M_2| \) is a Néel vector describing the staggered sublattice magnetization, \( M = |M_1 + M_2| \) is the total magnetization of the material, and \( P \) is the magnitude of the ferroelectric polarization along \( \hat{z} \) [the cubic (111) and equivalent directions in BiFeO$_3$]. This free energy is believed to capture the most relevant physics of BiFeO$_3$ bulk crystals, while for films, the dominant coupling between magnetic and ferroelectric orders is altered (see below).

If the Lifshitz term had been omitted, the AFM and FE orders would be decoupled, leading to a simple ground state that is a commensurate, isotropic Heisenberg antiferromagnet with \( |L_0|^2 = \frac{a}{c} \), plus an easy-axis ferroelectric with polarization \( P_0 = P_0 \hat{z}, P^2_0 = -a/u \). Clearly \( A < 0 \) and \( r > 0 \) for an antiferromagnetic ground state, while \( a < 0 \) and \( a_{\perp} > 0 \) for a ferroelectric ground state. The Lifshitz term induces a cycloidal ordering: the antiferromagnetic moment rotates in an arbitrary plane including \( \hat{z} \). It also increases the magnitude of \( L_0 \) and \( P_0 \). For a reference ground state about which to consider small perturbations, we take a cycloid with

\[ L_0(x) = L_0 \left[ \cos(qx) \hat{z} + \sin(qx) \hat{x} \right]. \] (2)

The cycloid direction is parallel to \( \hat{x} \) and the pitch is \( q = \alpha P_0/c' \). The strength of antiferromagnetic and polar orders are given by

\[ L_0^2 = -A + \frac{\alpha^2 P_0^2}{c'}, \quad P_0^2 = -a + \frac{\alpha^2 L_0^2}{u}. \] (3)

The linearized equations of motion for the ferroelectric-antiferromagnet order parameters include the familiar Landau-Lifshitz equations, complemented by the Debye equation of motion for the ferroelectric order parameter:

\[ \frac{\partial^2 L}{\partial t^2} = -r(\gamma L_0)^2 \left[ \frac{\delta F}{\delta L} - \left( \frac{\delta F}{\delta L_0} \right) L_0 \right], \] (4)

\[ \frac{\partial^2 P}{\partial t^2} = -f \frac{\delta F}{\delta P}. \] (5)

The constant \( \gamma \) is a gyromagnetic ratio with dimensions of \( (sG)^{-1} \), while \( f \) has dimensions of \( s^{-2} \) and plays a similar role in the ferroelectric equation of motion.

Details of the solution of these equations can be found elsewhere, along with a description of their coupling to external electromagnetic fields; in this work we would like to concentrate on the consequences for the prospective applications discussed in the Introduction. The main result of interest for applications is that an oscillating electric field at zero wavevector can couple to spin waves at nonzero wavevector, as a consequence of the cycloidal order present in the ground state. At lowest order and with a perfectly harmonic cycloid, the AC electric susceptibility has poles at the following shifted phonon and antiferromagnetic frequencies:

\[ \Omega_{PH}^2 = \frac{a}{\xi} + 2c' q^2, \] (6)

\[ \Omega_{AFM}^2 = 2c' q^2 - \frac{2(\alpha q L_0)^2}{a_{\perp}}, \] (7)

where contributions of order \( O(\xi q^2 / a_{\perp})^2 \) were dropped. In these formulas, we have defined \( \xi = r(\gamma L_0)^2 / f \approx 10 \) in BiFeO$_3$ and the AC electric field is \( Ee^{-i\omega t} \). The primed frequencies are defined in units of \( (\gamma L_0 / \sqrt{\tau}) \sim 10^{12} \) rad/s.

There are additional magneto-optical resonances that appear if a more realistic theory is constructed by including, for example, anharmonicity in the cycloidal ground state. The possible impact of these optically active spin waves for applications can be simply stated: the cycloidal structure of the material means that even zero-wavevector perturbations, which are potentially simpler to generate than the stripe-line-launched waves in current experiments, can couple to nonzero-wavevector spin waves with a preferred direction. Since antiferromagnetic spin waves generically have large group velocity even at large wavelength, this may provide a viable means for efficient launching of spin waves with electrical fields.

The cycloidal nature of the magnetic order leads to yet another potentially useful property for applications: the lowest frequency branch for spin wave propagation is strongly anisotropic. This occurs due to the presence of a non-trivial symmetry operation, namely we may rotate...
the cycloid pitch $q$ out of the $xz$ plane with no energy cost. This property implies the presence of a zero frequency mode with dispersion independent of $k_y^2$:

$$\omega^2 \approx c' \left[ \frac{k_x^2 + k_z^2}{8 q^2} - \frac{k_y^2}{q^2} \right],$$

(8)

where we dropped contributions $O(k^0)$. This soft mode dispersion is strongly anisotropic, and may be useful for electrical control of the spin wave group velocity via electrical switching of the $P_\parallel$ direction. Note that the spin wave group velocity is quite fast for propagation along the $xz$ plane, which is here defined by the polarization $P_0$ and the cycloid direction $q$. For this case the group velocity is given by $\gamma L_0 \sqrt{\tau c} \sim 10^8$ cm/s. Interestingly, the anisotropic dispersion found in Eq. (8) leads to zero group velocity (at $k \to 0$) for propagation along the $y$ direction (perpendicular to the cycloid plane).

The next section discusses the surprisingly different physics that occurs in BiFeO$_3$ films, where there is a uniform canted antiferromagnetic ground state rather than a cycloid, in order to predict how electrical switching of the magnetization direction will modify the spin-wave propagation.

III. SPIN-WAVE SWITCHING IN BIFEO$_3$ FILMS

The cycloid order is destroyed in epitaxially constrained BiFeO$_3$ films, as evidenced by the measurement of a much larger magnetic susceptibility and confirmed by neutron scattering experiments. For this reason, the Lifshitz invariant assumed in Eq. (11) can not be present or is too weak to produce noticeable effects (for example, the cycloid period in films is probably much larger than the typical domain size). Nevertheless, the films seem to have a weak macroscopic magnetization, of the order of 0.02$\mu_B$/Fe. This occurs even in the absence of applied fields and can only be explained by canting of the sublattice magnetizations. These considerations suggest the following model free energy for BiFeO$_3$ films:

$$F = \frac{a P_z^2}{2} + \frac{u P_x^4}{4} + \frac{a_\perp (P_x^2 + P_y^2)}{2} - P \cdot E$$

$$+ \sum_{j=1,2} \left[ \frac{r M_j^2}{2} + \frac{G M_j^4}{4} + c' \sum_i (\nabla M_j)_{ij}^2 \right]$$

$$+ J M_1 \cdot M_2 + d P \cdot M_1 \times M_2.$$  

(9)

Here $M_1$ and $M_2$ are sublattice magnetizations, and $P$ is the ferroelectric polarization. We assumed that canting occurs due to a linear magnetoelastic effect through a Dzyaloshinskii-Moriya vector equal to $d P$. As a result, this free energy is invariant under inversion at a center located in between the two sublattices. This coupling is expected if the low-temperature phase is imagined as resulting from symmetry breaking in a high-temperature cubic phase, but another possibility has been suggested based on electronic structure calculations. That work finds that the dominant contribution to the Dzyaloshinskii-Moriya vector is independent of $P$. In this case, the ferromagnetic vector $M$ is nearly decoupled from $P$. Experimental tests of how polarization switching modifies the magnetic structure in films are a high priority; tests so far indicate that the antiferromagnetic vector $L$ switches to remain perpendicular to $P$, but the response of $M$ has not been probed yet.

The reference ground state for film structures is an uniform-canted state,

$$M_{01} = M_0 (\sin \beta \hat{x} + \cos \beta \hat{y}),$$

(10a)

$$M_{02} = -M_0 (\cos \beta \hat{x} + \sin \beta \hat{y}),$$

(10b)

with canting angle approximately given by $\tan \beta \approx d P_0/(2J)$. The spin waves for this canted state were calculated a long time ago for propagation along the symmetry axis ($P_0$ in our case), perpendicular to the canting plane. There are two different spin wave modes: One is a low frequency mode, with linear dispersion $\tilde{\omega} = \sqrt{2 J} \tilde{k}$, where here $\tilde{\omega} = \omega/(\gamma M_0)$. The other is a high frequency mode, with gap equal to the Dzyaloshinskii-Moriya coupling, $\tilde{\omega} \approx \tilde{d} P_0$.

We now discuss the possibility of electronic excitation of spin waves in a canted multiferroic such as BiFeO$_3$ films. The canted-uniform magnetic order of the films imply that an electrical probe with wavevector $k$ will only excite spin waves at the same wavevector $k$. This is in drastic contrast to the case of bulk BiFeO$_3$ described above, where the cycloidal magnetic order allowed the transduction of a $k = 0$ excitation into an $nq$ excitation.

However, the presence of a macroscopic weak magnetization in the film state allows a whole new set of magnetoelectric functionalities. Below we provide a summary of our results, with a focus on electrical control of magnon propagation. A detailed description is available elsewhere. For example, the fact that the weak magnetization scales linearly with the electric polarization ($2M_0 \sin \beta \approx d P_0 M_0/J$) allows electrical excitation of low frequency spin waves provided the AC electric field is polarized along the weak magnetization direction ($x$ direction in our case). Moreover, an electric field polarized in the $yz$ plane (perpendicular to the weak magnetization) will excite a high frequency magnon. The latter effect may be observed optically in the far infrared region, since this will lead to an electromagnon resonance (a pole in the dielectric function) at the Dzyaloshinskii-Moriya frequency.

Moreover, the presence of a macroscopic weak magnetization will also lead to a sizable anisotropy in the low frequency magnon dispersion. The physical origin of this anisotropy is completely different than the one occurring in bulk BiFeO$_3$ [See Eq. (8) above]. In films, the presence of a weak magnetic moment implies the spin waves will produce a macroscopic AC magnetization with finite amplitude $\delta M$. From Maxwell’s equations we see that
this must create a magnetostatic field of the form
\[ h = -4\pi (\delta M \cdot \hat{n}) \hat{r}e^{i(k \cdot r - \omega t)}, \]
where \( \hat{n} \) is the spin wave propagation direction, \( \mathbf{k} = k \hat{n} \). The magnetostatic field contributes a term \( 2\pi (\delta M \cdot \hat{n})^2 \) to the free energy, tending to increase the spin wave frequencies whenever the quantity \( \delta M \) has a finite projection along \( \hat{n} \). As a consequence, the low frequency mode becomes anisotropic according to
\[ \omega^2(k) \approx 2Jc' \left( 1 + \frac{4\pi}{J} n^2 \right) k^2 + \frac{4\pi(dP_0)^2}{J} n^2. \] (12)

Interestingly, a gap of the order of \( \sqrt{4\pi/J(dP_0)} \) appears for spin waves propagating along the \( y \) direction (the direction of the Néel vector in our case). Similarly to the case of bulk BiFeO₃, this effect may be used as an electrical switch for spin waves propagating along the \( xz \) direction. Changing the orientation of \( P_0 \) electrically will drastically reduce the group velocity of a spin wavepacket propagating along the \( xz \) plane.

The physical origin of this gap lies in the anticrossing between the photon dispersion (\( \omega = ck \)) and the low frequency magnon dispersion, a phenomenon analogous to the formation of polaritons for optical phonon excitations. The magnetostatic propagation anisotropy Eq. (12) arises precisely because the spin waves travel with finite \( k > \omega/c \), which is always greater than a few \( \text{cm}^{-1} \) due to the small domain sizes. For this reason, the magnon dispersion is effectively gapped, but we emphasize that the strict \( k \rightarrow 0 \) limit, observable only at very long wavelengths (several centimeters) has \( \omega \rightarrow 0 \) and no orientation dependence as expected by symmetry.

### IV. CONCLUSIONS

In summary, we discussed the possibility of electrical excitation and control of spin waves in BiFeO₃ monocrystals and heterostructures. The presence of the inhomogeneous cycloidal order in bulk BiFeO₃ implies an AC electric field at \( k \approx 0 \) will excite spin waves at integer multiples of the cycloid wavevector (at \( k = nq \)). This shows that bulk BiFeO₃ is potentially useful as a source for finite wavevector magnons, which may become a viable alternative to the current strip-line designs. Although BiFeO₃ films do not have this functionality, spin waves may still be excited electrically, due to the presence of a Dzyaloshinskii-Moriya linear magnetoelectric effect.

We remark that changing the direction of the ferroelectric polarization may force a switch of the Néel vector orientation in a special class of multiferroic materials, an effect already demonstrated in BiFeO₃ films. In our work we showed in addition that BiFeO₃ monocrystals as well as heterostructures possess a sizable anisotropy in the lowest frequency spin wave dispersion. The combination of these two physical phenomena implies that the magnon group velocity may be stopped electrically in BiFeO₃, an important new functionality for devices based on spin wave propagation. Similar effects may be demonstrated in other well studied multiferroic materials, such as TbMnO₃,26 or BaNiF₃.27

The authors acknowledge useful conversations with J. Orenstein and R. Ramesh. This work was supported by WIN (RdS) and by NSF DMR-0238760 (JEM).

---

* Current address: Department of Physics and Astronomy, University of Victoria, Victoria, BC V8W 3P6, Canada.

1. M.P. Kostylev, A.A. Serga, T. Schneider, B. Leven, and B. Hillebrands. Spin-wave logical gates. Appl. Phys. Lett. 87, 153501 (2005).

2. A. Khitun and K. L. Wang. Nanoscale computational architectures with spin-wave bus. Superlattices and Microstructures 38, 184 (2005).

3. T. Zhao, A. Scholl, F. Zavaliche, K. Lee, M. Barry, A. Doran, M. P. Cruz, Y. H. Chu, C. Ederer, N. A. Spaldin, R. R. Das, D. M. Kim, S. H. Baek, C. B. Eom and R. Ramesh. Electrical control of antiferromagnetic domains in multiferroic BiFeO₃ films at room temperature. Nature Materials 5, 823 (2006).

4. M.E. Lines and A.M. Glass. Principles and Applications of Ferroelectricity and Related Materials. Oxford, U.K. (1977).

5. V.G. Bar’yakhtar and I.E. Chupis. Phenomenological theory of a ferroelectric magnet. Sov. Phys. Solid State 10, 2818 (1969); V.G. Bar’yakhtar and I.E. Chupis. Quantum theory of oscillations in a ferroelectric ferrimagnet. Sov. Phys. Solid State 11, 2628 (1970).

6. D.R. Tilley and J.F. Scott. Frequency dependence of magnetoelectric phenomena in BaMnF₄. Phys. Rev. B 25, 3251 (1982).

7. R. Ramesh and N. A. Spaldin. Multiferroics: Progress and prospects in thin films. Nature Materials 6, 21 (2007).

8. R. de Sousa and J.E. Moore. Optical coupling to spin waves in the cycloidal multiferroic BiFeO₃. cond-mat/0706.1260, Phys. Rev. B in press.

9. A. Sparavigna, A. Strigazzi, and A. Zvezdin. Electric-field effects on the spin-density wave in magnetic ferroelectrics. Phys. Rev. B 50, 2953 (1994).

10. I. Sosnowska, T. Peterlin-Neumaier, and E. Steichele. Spiral magnetic ordering in bismuth ferrite. J. Phys. C: Solid State Phys. 15, 4835 (1982).

11. A.V. Zalesskii, A.K. Zvezdin, A.A. Frolov, and A.A. Bush. 57Fe NMR study of a spatially modulated magnetic structure in BiFeO₃. JETP Lett. 71, 465 (2000).

12. B. Ruegg, S. Zvyagin, A. Pyatakov, A. Bush, J. F. Li, V. I. Belotelov, A. K. Zvezdin, and D. Viehland. Magnetic-field-induced phase transition in BiFeO₃ observed by high-field electron spin resonance: Cycloidal to homogeneous spin order. Phys. Rev. B 69, 064114 (2004).

13. F. Bai, J. Wang, M. Wuttig, J.F. Li, N. Wang, A. Pyatakov, A. Zvezdin, L.E. Cross, D. Viehland. Destruction of spin cycloid in (111) c-oriented BiFeO₃ thin films by
epitaxial constraint: enhanced polarization and release of latent magnetization. Appl. Phys. Lett. 86, 032511 (2005).
14. H. Béa, M. Bibes, S. Petit, J. Kreisel, and A. Barthélémy. Structural distortion and magnetism of BiFeO3 epitaxial thin films: A Raman spectroscopy and neutron diffraction study. Phil. Mag. Lett. 87, 165 (2007).
15. D. L. Fox and J. F. Scott. Ferroelectrically induced ferromagnetism. J. Phys C 10, L329 (1977).
16. C. Ederer and N. A. Spaldin. Weak ferromagnetism and magnetoelastic coupling in bismuth ferrite. Phys. Rev. B 71, 060401(R) (2005)
17. G.F. Herrmann. Orientation dependence of resonance second harmonic, and spin waves in anisotropic ferromagnetic crystals. J. Phys. Chem. Solids 25, 347 (1964).
18. R. de Sousa and J.E. Moore. Electrical control of magnon propagation in multiferroic BiFeO3 films. Appl. Phys. Lett. in press.
19. M. Covington, T.M. Crawford, and G.J. Parker. Time-Resolved Measurement of Propagating Spin Waves in Ferromagnetic Thin Films. Phys. Rev. Lett. 89, 237202 (2002).
20. T. Kimura, T. Goto, H. Shintani, K. Ishizaka, T. Arima, and Y. Tokura. Magnetic control of ferroelectric polarization. Nature 426, 55 (2003).
21. C. Ederer and N.A. Spaldin. Electric-field-switchable magnets: The case of BaNiF4. Phys. Rev. B 74, 020401(R) (2006).