Influence of electric fields on spin waves in simple ferromagnets: Role of the flexoelectric interaction

D. L. Mills and I. E. Dzyaloshinskii
Department of Physics and Astronomy, University of California–Irvine, Irvine, California 92697, USA
(Received 11 August 2008; published 18 November 2008)

We discuss the influence of electric fields on spin waves in simple ferromagnets produced by the flexoelectric interaction. Such terms produce a frequency shift that is linear in both the applied electric field and the wave vector of the spin waves. Above a certain critical electrical field, ferromagnetism is unstable. We discuss the nature of the conical cycloidal state that appears above the critical field for a ferromagnet with uniaxial anisotropy and exchange.

DOI: 10.1103/PhysRevB.78.184422 PACS number(s): 75.30.Ds, 75.40.Gb

I. INTRODUCTION

An issue of fundamental interest is the influence of electric fields on magnetically ordered materials. Interest in such phenomena is very active currently because of potential applications to devices.

Many years ago, it was noted that magnetoelectric phenomena are present in materials within which both time-reversal symmetry and parity are not good symmetry operations.[1] Since then, magnetoelectric phenomena of diverse sorts have been studied both in ferromagnets and antiferromagnets. For instance, Rado and co-workers[2] observed an electric-field-induced shift in the ferromagnetic resonance frequency of lithium ferrite. In a recent discussion, Sousa and Moore[3] presented a theoretical description of electric-field effects on spin waves in the multiferroic material $BeFeO_3$.

It is of interest to inquire if electric fields can influence the properties of simple ferromagnets, where inversion symmetry is present. While electric fields cannot couple to the uniform magnetization in such materials, they can interact with its spatial gradients. The first prediction of the influence of the effect of an electric field on the properties of simple ferromagnets was presented some decades ago when it was argued that in the itinerant 3*d* ferromagnets, application of an electric field will shift the frequency of finite wave-vector spin waves by virtue of the Doppler shift associated with the drift velocity imparted to the moment bearing 3*d* electrons by the electric field.[4] Very recently, the experimental measurement of such a Doppler shift has been reported,[5] with magnitude rather close to that predicted. Subsequent authors have presented more complete theoretical descriptions of this effect.[6,7]

It is the case that couplings one may refer to as flexoelectric interactions are also present in simple ferromagnets, as noted by Bar'yakhtar *et al.*[8] These also couple electric fields to gradients in the magnetization and are necessarily present in insulating as well as metallic ferromagnets. Their presence results in a nonzero electric polarization within domain walls of Neèl character,[8] and they also influence materials in which the magnetization is spatially varying, such as spiral magnets.[9]

One of us has demonstrated that in the presence of sufficiently strong electric fields applied parallel to the magnetization, flexoelectric interactions render the uniform ferromagnetic state unstable with respect to nucleation of a Neèl domain wall.[10] In the present paper, we explore the influence of this coupling on spin waves in ferromagnets. We find a frequency shift that is linear in both the wave vector of the spin wave, and the applied electric field. This is, thus, a frequency shift very similar in nature to the Doppler shift mentioned above. However, the Doppler shift assumes its maximum value when the electric field and wave vector of the spin wave are parallel to each other and vanishes when they are perpendicular. In contrast, the flexoelectric induced shift is maximum when the electric field and wave vector are perpendicular to each other and to the easy axis, and it vanishes when the two are parallel. Thus, it will be possible to distinguish experimentally between the two sources of frequency shift. A measurement of the flexoelectric induced shift would be most valuable since it would provide one with information on the magnitude of the phenomenological coupling constants which that the theory.

We also show that application of a sufficiently large electric field perpendicular to the magnetization renders the uniform ferromagnetic state unstable. This critical field is larger by a factor of π than the critical field discussed in Ref. 10. For a uniaxial ferromagnet, in the absence of an applied Zeeman field, the new high-field state is cycloidal in character, with the magnetizaion in the plane which contains the electric field. This plane is perpendicular to the easy axis. Application of an external Zeeman field along the easy axis leads to a conical cycloidal state with net magnetization along the easy axis. We note that in Ref. 11, the role of flexoelectric terms in producing a similar state in the antiferromagnet $BiFeO_3$ was discussed. However, $BiFeO_3$ does not have inversion symmetry; the point we wish to make in the present paper is that even in the simplest ferromagnets where inversion symmetry is present, sufficiently strong electric fields can drive a transition to a conical cycloidal state.

In Sec. II, we introduce the flexoelectric couplings and discuss their influence on spin waves. We note in this section that ferromagnetism is unstable above a stated critical field. Section III explores the character of the new high-field state, and concluding remarks are in Sec. IV.

II. SPIN WAVES

We write the flexoelectric coupling as $V_E=V_E^{(1)}+V_E^{(2)}$, where

$$V_E^{(1)} = b_1 \int d^3r[\vec{E}\cdot\vec{M}(\vec{r})][\vec{\nabla}\cdot\vec{M}(\vec{r})], \tag{1a}$$

and

$$V_E^{(2)} = b_2 \int d^3r\vec{E}\cdot\{\vec{M}(\vec{r})\times[\vec{\nabla}\times\vec{M}(\vec{r})]\}. \tag{1b}$$

Here b_1 and b_2 are phenomenological constants. We shall assume we have a ferromagnet with an easy axis parallel to $\hat{z}$, and we shall take the spatially uniform electric field to lie in the xz plane. Thus $\vec{E}=E_\perp\hat{x}+E_\parallel\hat{z}$. We then find

$$V_E^{(1)} = b_1E_\parallel \int d^3rM_z\left[\frac{\partial M_x}{\partial x}+\frac{\partial M_y}{\partial y}\right] + b_1E_\perp \int d^3rM_x\left[\frac{\partial M_y}{\partial y}+\frac{\partial M_z}{\partial z}\right], \tag{2a}$$

and

$$V_E^{(2)} = -b_2E_\parallel \int d^3r\left[M_x\frac{\partial M_z}{\partial x}+M_z\frac{\partial M_x}{\partial z}\right] - b_2E_\perp \int d^3r\left[M_y\frac{\partial M_x}{\partial y}+M_z\frac{\partial M_x}{\partial z}\right]. \tag{2b}$$

We have used the identity $\int d^3rM_\alpha(\partial M_\alpha/\partial x_\beta) =(1/2)\int d^3r\partial M_\alpha^2/\partial x_\beta=0$ to cast the results in the form displayed in Eq. (2).

To explore that nature of spin waves, we use as the Hamiltonian the form $H=H_0+V_E$, where H_0 contains exchange, anisotropy, and dipole interaction terms. In this section we shall not require the precise form of H_0. We shall obtain the equation of motion of the magnetization density operator M_α.

This is given by $i\hbar\dot{M}_\alpha=[M_\alpha,H]$, and one may generate the various contributions to the equations of motion through use of the commutator $[M_\alpha(\vec{r}),M_\beta(\vec{r}')]=i\mu_0\varepsilon_{\alpha\beta\gamma}M_\gamma(\vec{r})\delta(\vec{r}-\vec{r}')$. Here μ_0 is the magnetic moment in each unit cell of the crystal. After the commutators are evaluated, the equations of motion for M_x and M_y are then linearized in the spirit of spin-wave theory.

After linearization, one finds the following simple result:

$$[M_{x,y}(\vec{r}),V_E] = -i\mu_0bE_\perp M_S\frac{\partial M_{x,y}}{\partial y}, \tag{3}$$

where $b=b_1+b_2$. After the equations of motion are generated, one then seeks plane-wave solutions of the form $M_{x,y}(\vec{r})=m_{x,y}\exp(i\vec{k}\cdot\vec{r}-i\Omega t)$ to find

$$\tilde{\Omega}m_{x,y} = \{m_{x,y},H_0\}, \tag{4}$$

where the symbol $\{m_{x,y},H_0\}$ stands for the terms in the equations of motion generated from H_0, $\tilde{\Omega}=\Omega-\gamma bE_\perp M_Sk_y$, and $\gamma=\mu_0/\hbar$ is the gyromagnetic ratio.

It then follows, independently of the detailed structure of H_0, that the spin-wave frequency in the presence of the electric field is given by

$$\Omega_E(\vec{k}) = \Omega_0(\vec{k}) + \gamma bE_\perp M_Sk_y, \tag{5a}$$

a result we also write as

$$\Omega_E(\vec{k}) = \Omega_0(\vec{k}) - \gamma b\vec{k}\cdot[\vec{E}\times\vec{M}_S]. \tag{5b}$$

As discussed in the introduction, the flexoelectric terms lead to a term in the dispersion relation linear in the wave vector. We see that in contrast to the Doppler shift present in the metallic ferromagnets, the coefficient of the linear term has its maximum value when the propagation direction of the spin wave is perpendicular to the electric field (and also perpendicular to the easy axis). We recall that the Doppler shift $\delta\Omega_D(\vec{k})$ is given by $\delta\Omega_D(\vec{k})=\vec{v}_d\cdot\vec{k}$, where the drift velocity of the moment bearing carriers $\vec{v}_d$ is parallel to the electric field, which drives the current in simple ferromagnets such as considered here. Estimates of the mobility of the moment bearing electrons are found in Ref. 4. The estimate in this paper provides a Doppler shift within a factor of 2 of that observed in permalloy by the authors of Ref. 5.

To proceed further, we require a specific form for $\Omega_0(\vec{k})$. We follow Ref. 10 by choosing

$$H_0 = \int d^3r\left[\frac{A}{2}\sum_\alpha|\vec{\nabla}M_\alpha|^2-\frac{a}{2}M_z^2-h_0M_z\right]. \tag{6}$$

The z axis is, thus, the easy axis, and we have added a Zeeman field h_0 parallel to the easy axis. We then have

$$\Omega_E(\vec{k}) = \gamma\{h_0+aM_S+AM_sk^2-bM_SE_\perp k_y\}. \tag{7}$$

We have chosen the sign of $E_\perp$ so that the linear term has negative slope when $k_y>0$.

If we chose the wave vector in the y direction, then the dispersion curve has a minimum shifted away from $k_y=0$ by the amount $k_m=bE_\perp/2A$; the minimum value of the spin-wave frequency is given by

$$\Omega_m = \gamma\left\{h_0+aM_s-\frac{b^2E_\perp^2M_S}{2A}\right\}. \tag{8}$$

At a certain critical electric field $E_\perp^{(c)}=\frac{2[A(a+h_0/M_S)]^{1/2}}{b}$, the frequency Ω_m is driven to zero. We provide a schematic illustration of the nature of the dispersion relation in Fig. 1. For applied electric fields greater than this critical field, we have spin waves with negative excitations energies, and the ferromagnetic state is unstable. In Sec. III, we will describe the new high-field state.

It is interesting to compare the critical field above with that discussed in Ref. 10, where it was demonstrated that application of an electric field parallel to the magnetization will produce an instability of the uniform ferromagnetic state, with the high-field state one in which a Neèl wall forms spontaneously. When we set $h_0=0$, the critical field described in this paper is larger than that in Ref. 10 by a factor of π.

We remark that if our simple ferromagnet has a crystal structure which admits optical phonons, then in the presence of an electric field there also will be linear terms in the wave vector in their dispersion relation. This is a direct consequence of the fact that the presence of the spontaneous mag-

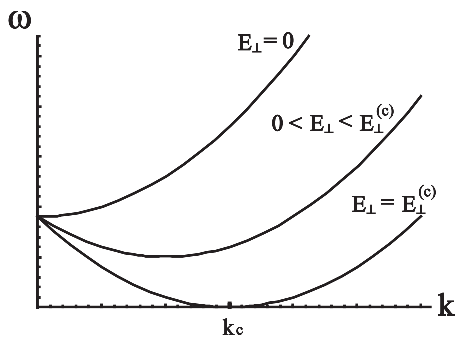

FIG. 1. A schematic illustration of the effect of the flexoelectric terms on the spin-wave dispersion relation in the ferromagnetic state. The wave vector is in the y direction, perpendicular to the magnetization, and the electric field $E_\perp$ is perpendicular to the x axis. In the absence of the electric field, $E_\perp=0$, we have the classical dispersion relation quadratic in wave vector, with a gap produced by the combination of the anisotropy and Zeeman field. The electric field leads to a linear term in wave vector, with negative slope and a minimum in the dispersion curve at a nonzero value of the wave vector. When the electric field is increased to the critical value above which ferromagnetism is unstable, we have a "soft spin wave" at the wave vector k_c.

netization breaks time-reversal symmetry, while application of an electric field leads to a breakdown of inversion symmetry. Thus, in the presence of an electric field $\Omega_{\text{opt}}(\vec{k}) \neq \Omega_{\text{opt}}(-\vec{k})$, just as for spin waves. This asymmetry is brought about by two kinds of terms in the energy: the standard magnetoelectric term, which mixes the oscillating electric polarization $\vec{p}$ of the optical phonon, and the magnetic polarization $\vec{m}$ of the spin waves. These terms have the form $C_{\text{me}}^{(1)}(\vec{M}\cdot\vec{m})(\vec{E}\cdot\vec{p})+C_{\text{me}}^{(2)}(\vec{M}\cdot\vec{p})(\vec{E}\cdot\vec{m})$, and then there are flexoelectric terms analogous to those in Eq. (1): $C_{\text{flex}}^{(1)}\vec{M}\cdot(\vec{p}\times[\vec{\nabla}\times\vec{m}])+C_{\text{flex}}^{(2)}(\vec{M}\cdot\vec{m})\vec{\nabla}\cdot\vec{p}$. In these expressions, $\vec{M}$ is the static magnetization. Straightforward but lengthy calculations produce the linear terms in wave vector in the optical phonon spectra. The expressions are complex and involve the several coefficients just displayed; at this point there is little motivation to present the analysis or final expressions. However, it may be possible to observe the electric-field-induced linear terms in the optical phonon-dispersion relation directly by a technique such as the Raman scattering of light, so we wish to point out the existence of these terms.

III. HIGH-FIELD STATE

In this section, with electric field applied in the x direction, we suppose that all the magnetization components M_α depend only on the coordinate y. Then upon noting the identity $\int dy M_x(\partial M_y/\partial y) = -\int dy M_y(\partial M_x/\partial y)$, we may arrange the expression for V_E to have the form

$$V_E = \frac{bE_\perp}{2}\int d^3r\left[M_x\frac{\partial M_y}{\partial y} - M_y\frac{\partial M_x}{\partial y}\right]. \tag{9}$$

We seek extrema of the total energy after writing the magnetization components in the form $M_x=M_S\sin\theta(y)\cos\varphi(y)$, $M_y=M_S\sin\theta(y)\sin\varphi(y)$, and $M_z=M_S\cos\theta(y)$. The energy per unit area e of this state may then be written as $e=M_S^2\int dy U(y)$, where, after using the energy of the uniform ferromagnetic states as the zero of energy, we have

$$U(y) = \frac{A}{2}\left\{\left(\frac{\partial\theta}{\partial y}\right)^2 + \sin^2\theta\left(\frac{\partial\varphi}{\partial y}\right)^2\right\} + \left\{\frac{bE_\perp}{2}\sin^2\theta\frac{\partial\varphi}{\partial y} + \frac{a}{2}\sin^2\theta + 2\frac{h_0}{M_S}\sin^2\frac{\theta}{2}\right\}. \tag{10}$$

We minimize this energy for a state in which $\theta(y)=\theta_0$ and $\varphi(y)=k_E y$, where θ_0 and k_E are independent of y. One finds

$$k_E = -\frac{bE_\perp}{2A}, \tag{11a}$$

and

$$\cos\theta_0 = \frac{h_0}{h_0 + h_c(E_\perp)}, \tag{11b}$$

where

$$h_c(E_\perp) = \frac{b^2M_S}{4A}\{E_\perp^2 - (E_\perp^c)^2\}. \tag{11c}$$

In Eq. (11c), $E_\perp^{(c)}$ is the critical field introduced in Sec. II, above which ferromagnetism is unstable. We assume in this section that $E_\perp > E_\perp^{(c)}$. The energy density U associated with this state is given by

$$U = -\frac{1}{2M_S}\frac{h_c(E_\perp)^2}{h_0 + h_c(E_\perp)}. \tag{11d}$$

In the absence of an applied Zeeman field ($h_0=0$), we have a cycloidal state wherein the magnetization lies in the plane perpendicular to the easy axis. Application of a Zeeman field parallel to the easy axis tilts the magnetization out of plane, and we have then a conical cycloidal state. The sense of rotation of the transverse components of the spin, as one looks down on the xy plane from above, is controlled by the sign of the parameter b for $E_\perp$ applied in the $+x$ direction. As the Zeeman field is increased, we achieve nearly ferromagnetic alignment of the spins when h_0 is large compared to $h_c(E_\perp)$.

The phase transition considered above is a type of commensurate-incommensurate (CI) transition: the commensurate state with magnetization parallel to the easy axis changes to the conical cycloidal wave with the finite wave vector k_E given by Eq. (11a). On the other hand, the CI considered in Ref. 10 belongs to a different type of CI (Ref. 12): when the electric field reaches the critical value E_t a single Neèl domain wall appears. As the electric field increases, a lattice of domain walls develops, and these evolve with further increase in the field to a simple cycloidal wave.

IV. FINAL COMMENTS

In Ref. 10, it was demonstrated that in the presence of an electric field applied parallel to the easy axis, the ferromagnetic state is unstable with respect to nucleation of a Neèl wall in the presence of flexoelectric coupling for electric fields larger than a certain critical field. We have shown here that a field applied perpendicular to the easy axis also renders ferromagnetism unstable; with Zeeman field absent, the transverse critical field is larger than the longitudinal critical field by a factor of π for the particular model of an easy axis ferromagnetic examined in the two papers. For electric fields below the transverse critical field, we find the flexoelectric coupling produces a term linear in wave vector in the spin-wave dispersion relation so there is an off-center minimum in the dispersion relation for propagation in the plane perpendicular to the easy axis, and also perpendicular to the in plane component of the applied electric field.

A measurement of the linear term in the dispersion relation in the low-field state would be of great importance because this would supply us with the value of the central parameter of the theory, the flexoelectric coupling constant $b=b_1+b_2$. At the time of this writing, we have no knowledge of the value of this parameter in any material. The dependence of the spin-wave dispersion relation on electric field offers direct access to this important parameter. It would be of considerable importance for such a measurement to be carried out.

A geometry which could be utilized is readily realized in the laboratory. One can envision a film magnetized in plane, with the wave vector of the spin wave also in plane and perpendicular to the easy axis. An electric field perpendicular to the film surfaces may be generated through suitable electrodes on the film surfaces. It is probable that the frequency shift will not be large. In this regard, the measurements reported in Ref. 4 are of great interest. The frequency shift produced by the Doppler effect is quite small, 0.3%, but nonetheless it is clearly measurable.

ACKNOWLEDGMENT

The research of D.L.M. was supported by the U.S. Army through Contract No. CS000128

[1] I. E. Dzaloshinskii, Sov. Phys. JETP **10**, 628 (1960).
[2] G. T. Rado, C. Vittoria, J. M. Ferrari, and J. P. Remeika, Phys. Rev. Lett. **41**, 1253 (1978).
[3] Rogerio de Sousa and Joel E. Moore, Appl. Phys. Lett. **92**, 022514 (2008).
[4] Pascal Lederer and D. L. Mills, Phys. Rev. **148**, 542 (1966).
[5] Vincent Vlaminck and Mattieu Bailleul, Science **322**, 410 (2008).
[6] Y. B. Bazaliy, B. A. Jones, and S. C. Zhang, Phys. Rev. B **57**, R3213 (1998).
[7] J. Fernández-Rossier, M. Braun, A. S. Nuñez, and A. H. MacDonald, Phys. Rev. B **69**, 174412 (2004).
[8] V. G. Bar'yakhtar, V. A. Lvov, and D. A. Yablonskii, JETP Lett. **37**, 673 (1983).
[9] M. Mostovoy, Phys. Rev. Lett. **96**, 067601 (2006).
[10] Igor Dzyaloshinskii, Europhys. Lett. **83**, 67001 (2008).
[11] A. Sparavigna, A. Strigazzi, and A. Zvezdin, Phys. Rev. B **50**, 2953 (1994).
[12] I. E. Dzyaloshinskii, Sov. Phys. JETP **20**, 665 (1965).