ISSN 1674-1056
http://cpb.iphy.ac.cn
http://iopscience.iop.org/cpb
中国物理 B
Chinese Physics
中国物理
Chinese Physics B

Dynamical anisotropic magnetoelectric effects at ferroelectric/ferromagnetic insulator interfaces

Yaojin Li(李耀进), Vladimir Koval, Chenglong Jia(贾成龙)
Citation: Chin. Phys. B . 2019, 28(9): 097501 . **doi:** 10.1088/1674-1056/28/9/097501
Journal homepage: http://cpb.iphy.ac.cn; http://iopscience.iop.org/cpb

What follows is a list of articles you may be interested in

Effect of magnetic properties of soft magnetic phase on the energy product of an exchange-spring magnet

Jia Li-Ying, Yin Jin-Hua, Ma Xing-Qiao
Chin. Phys. B . 2014, 23(2): 027501 . **doi:** 10.1088/1674-1056/23/2/027501

The effect of Fe doping on structural and magnetic properties of nanocrystallite $Co_2Cr_{1-x}Fe_xAl$ Heusler alloys prepared by mechanical alloying

M. Hakimi, P. Kameli, H. Salamati
Chin. Phys. B . 2012, 21(11): 117502 . **doi:** 10.1088/1674-1056/21/11/117502

The doping effects of $BiFe_{1-x}Co_xO_3$ (x=0.0-0.8) in layered perovskite $Bi_4Ti_3O_{12}$ ceramics

Yang Wen-Lu,Chen Chun-Yan,Mao Xiang-Yu,Chen Xiao-Bing
Chin. Phys. B . 2012, 21(4): 047502 . **doi:** 10.1088/1674-1056/21/4/047502

Excellent soft magnetic properties realized in FeCoN thin films

Zhang Lu-Ran,Lü Hua,Liu Xi,Bai Jian-Min,Wei Fu-Lin
Chin. Phys. B . 2012, 21(3): 037502 . **doi:** 10.1088/1674-1056/21/3/037502

SPECIAL TOPIC — 110th Anniversary of Lanzhou University

Dynamical anisotropic magnetoelectric effects at ferroelectric/ferromagnetic insulator interfaces*

Yaojin Li(李耀进)[1,2], Vladimir Koval[3], and Chenglong Jia(贾成龙)[1,†]

[1] Key Laboratory for Magnetism and Magnetic Materials of Ministry of Education, Lanzhou University, Lanzhou 730000, China
[2] Electronic Materials Research Laboratory International Center for Dielectric Research, Xi'an Jiaotong University, Xi'an 710049, China
[3] Institute of Materials Research, Slovak Academy of Sciences Watsonova 47, 04001 Kosice, Slovakia

(Received 7 June 2019; revised manuscript received 19 July 2019; published online 23 August 2019)

The interfacial magnetoelectric interaction originating from multi-orbital hopping processes with ferroelectric-associated vector potential is theoretically investigated for complex-oxide composite structures. Large mismatch in the electrical permittivity of the ferroelectric and ferromagnetic materials gives rise to giant anisotropic magnetoelectric effects at their interface. Our study reveals a strong linear dynamic magnetoelectric coupling which genuinely results in electric control of magnetic susceptibility. The constitutive conditions for negative refractive index of multiferroic composites are determined by the analysis of light propagation.

Keywords: interfacial magnetoelectric effect, ferromagnetic insulator, magnetic susceptibility, refractive index

PACS: 75.30.Cr, 75.50.Dd, 78.20.Ci **DOI:** 10.1088/1674-1056/ab37f7

1. Introduction

The study on the magnetic response of materials subjected to an external electric fields has generated great interest from both fundamental physics and potential application perspectives.[1–8] In particular, the direct coupling between electrical and magnetic polarization allows for purely electrical manipulation of magnetism with promising multifunctionality and low energy consumption. From the symmetry point of view, the magnetoelectric (ME) interactions can only develop in systems with both time-reversal and spatial-inversion symmetry breaking,[2] such as single phase magnetoelectric hexaferrites,[9] multiferroic bismuth ferrites[10] or ferroelectric/ferromagnetic (FE/FM) composite structures.[11] For single phase multiferroics, the mechanisms of ferroelectricity with spin origin have been identified[9,12] and the multiferroic response is well documented experimentally and theoretically.[13] On the other hand, a comprehensive understanding of coupling between spin and charge degrees of freedoms in the insulating interface state of composite multiferroics is still missing. In artificial FE/FM multiferroics, the studies on the interfacial ME effect have thus far remained interesting such as the electrostatic screening effect,[5,14–17] ionic liquid gating,[18–20] orbital reconstruction,[20–24] *etc*. However, this electric field effect can be ruled out in case of the FM insulator (FMI) interfaces due to the absence of mobile charge carriers.

In this paper, the non-local FE-associated vector potential is demonstrated to give rise to an anisotropic ME interaction through the multiorbital hopping occurring in FMI. The large electrical permittivity mismatch between FE and FMI is found to bring about a significant enhancement of the interfacial ME effects. The dynamic magnetic response driven by electromagnetic waves via strong linear ME coupling is investigated in detail. The electrical tuning of magnetic permeability in FE/FMI is discussed as well. Moreover, the negative refractive index is possible in the FM system with small electric permittivity. All these findings not only contribute to the current understanding of the magnetoelectric effects in composite structures but also fetch in a new perspective to nanoscale composite multiferroics.

2. Theoretical model

Let us consider lightly doped manganese oxides $La_{1-x}Sr_xMnO_3$ (LSMO with $0.1 \leq x \leq 0.15$) as the potential candidates of FMI in our setup.[25] The phase diagram of LSMO is well established[26,27] and the metallic $La_{1-x}Sr_xMnO_3$ ($0.175 < x < 0.48$) has been used for multiferroic heterosturctures.[28–32] A selected list of values for the magnetoelectric coupling constant of artificial multiferroic systems based on LSMO is given in Table 1.

*Project supported by the National Natural Science Foundation of China (Grant Nos. 11474138 and 11834005), the Fund from the Ministry of Science and Technology of China (Grant No. CN-SK-8-4), the Science Foundation from the Slovak Academy of Sciences (Grant No. 2/0059/17), and the Science Fund from the Slovak Research and Development Agency (Grant No. APVV SK-CN-2017-0004).

†Corresponding author. E-mail: cljia@lzu.edu.cn

© 2019 Chinese Physical Society and IOP Publishing Ltd

Table 1. Values of the magneto-electric coupling coefficient, α, reported in the literature for selected multiferroic heterostructures. T is the temperature and LSMO stands for $La_{1-x}Sr_xMnO_3$.

System	α/(s/m)	T/K	Coupling	FM material	Static/dynamic
LSMO ($x=0.33$)/BTO[a]	0.4–2.3×10^{-7}	200	strain	metal	static
LSMO ($x=0.3$)/PMN-PT[b]	6×10^{-8}	300	strain	metal	static
LSMO ($x=0.2$)/PZT[c,d]	6.2×10^{-9}	100	charge	metal	static
LSMO ($x=0.2$)/PZT[d]	−1.35×10^{-8}	180	charge	metal	static
LSMO ($x\in(0.1,0.15)$)/FE	$\sim 10^{-7}$	theory	electric field	insulator	dynamic

[a]Ref. [29]; [b]Ref. [30]; [c]Ref. [31]; [d]Ref. [32].

In these perovskite systems, the five (Mn) d-orbitals split under the octahedral crystal field into three lower t_{2g} and two upper e_g orbitals, the Jahn–Teller distortions further lift the double degeneracy of e_g orbitals.[33,34] The three t_{2g} electrons are mainly localised, whereas the e_g electrons are mobile and hopping between Mn ions through the bridge of O p-orbitals. All spins in the active shell are aligned parallel to each other by a strong Hund coupling. The ME properties can be obtained from the two-orbitals Kondo lattice model with large on-site Hubbard interactions[35–38]

$$\begin{aligned} H &= -\sum_{ij} t_{ij} c^{\dagger}_{i\sigma} c_{j\sigma} - J_{\mathrm{H}} \sum_{i} \left[\boldsymbol{S}_i \cdot (\boldsymbol{s}_{i\alpha} + \boldsymbol{s}_{i\beta}) + \boldsymbol{s}_{i\alpha} \cdot \boldsymbol{s}_{i\beta} \right] \\ &\quad + \sum_{i} \left[\Delta n_{i\beta} + U n_i (n_i - 1) \right], \end{aligned} \tag{1}$$

where α and β denote two e_g orbitals, respectively. $c_{i\sigma}$ is the destruction operator of an itinerant electron with spin $\boldsymbol{\sigma}$. t_{ij} describes the hoping between i and j sites, more precisely, between filled and empty orbitals at different sites, which is essential for the ME interaction. $J_{\mathrm{H}} > 0$ is the Hund coupling, $\boldsymbol{s}_{i\alpha} = \frac{1}{2} c^{\dagger}_{i\alpha} \boldsymbol{\sigma} c_{i\alpha}$ is the spin of the itinerant electron on the orbital α at site i, and the local spin $\boldsymbol{S}_i$ is approximated by a classical spin. $\Delta > 0$ is energy splitting between the two orbitals at site i, and $U > 0$ describes the Coulomb repulsion with $n_i = n_{i\alpha} + n_{i\beta}$ being the total number of itinerant electrons on the site. For $U = \Delta = 0$, an electron jumping between nearest-neighbor sites does not require energy (J_{H}), if all the spins are parallel. Hence, double exchange and FM ordering occur in the system.[40–42] On the other hand, the hoping terms involving only single-orbital per site with the limit of large U result in the t–J model with antiferromagnetic interaction.[43] Here, to second order perturbations of multi-orbital hopping, one gets an effective ferromagnetic interaction between the spins[38]

$$H_{\mathrm{FM}} = -\frac{t^2}{U'} (\boldsymbol{m}_i \cdot \boldsymbol{m}_j + 1), \tag{2}$$

where $U' = U + \Delta - J_{\mathrm{H}}/4$ and $\boldsymbol{m}_i$ is the unit vector along the local spin $\boldsymbol{S}_i$.

By interfacing FMI layer with FE films, the effective electric field develops in FMI. Taking into account the boundary conditions for the electric field at the interfaces between materials[39]

$$\hat{\boldsymbol{e}}_{\mathrm{n}} \cdot \left(\boldsymbol{D}^{\mathrm{FE}} - \boldsymbol{D}^{\mathrm{FM}} \right) = \sigma_{\mathrm{f}} \quad \text{and} \quad \hat{\boldsymbol{e}}_n \times \left(\boldsymbol{E}^{\mathrm{FE}} - \boldsymbol{E}^{\mathrm{FM}} \right) = 0, \tag{3}$$

where $\hat{\boldsymbol{e}}_n$ is a unit vector normal to the FE/FMI interface (hereafter we take $\hat{\boldsymbol{e}}_n \| \hat{\boldsymbol{e}}_z$, the z axis has its origin at the FMI interface), $\boldsymbol{D}^{\mathrm{FE}}$ and $\boldsymbol{D}^{\mathrm{FM}}$ are the displacement field in FE and FMI, respectively, the vanishing of free charges (*i.e.*, $\sigma_{\mathrm{f}} = 0$) at the insulator interfaces leads to

$$\boldsymbol{E}^{\mathrm{FM}}_z = \frac{\boldsymbol{D}^{\mathrm{FE}}_z}{\varepsilon_{\mathrm{FM}}} \quad \text{and} \quad \boldsymbol{E}^{\mathrm{FM}}_{x,y} = \boldsymbol{E}^{\mathrm{FE}}_{x,y}, \tag{4}$$

where $\varepsilon_{\mathrm{FM}}$ is the dielectric constant of FE and FMI. Considering $\boldsymbol{D}^{\mathrm{FE}} \sim 10^3 \varepsilon_0 \boldsymbol{E}$ with $\boldsymbol{E}$ being an applied electric field by gate voltage, the normal component of the effective electric field $\boldsymbol{E}^{\mathrm{FM}}_z$ in FMI is significantly enhanced up to several orders ($\varepsilon_{\mathrm{FM}}/\varepsilon_0 \sim 1$) compared with the other two in-plane components $\boldsymbol{E}^{\mathrm{FM}}_{x,y}$. As a consequence, giant anisotropic electric-field effects are expected in composite FE/FMI heterostructures.

In presence of electric field, the hopping amplitude can be modified in a gauge-invariant manner with the Peierls substitution,[44,45] $t_{ij} \to t_{ij} \exp\left[-\mathrm{i}e \int_{\boldsymbol{r}_i}^{\boldsymbol{r}_j} \mathrm{d}\boldsymbol{r} \cdot \boldsymbol{A}(\boldsymbol{r}, t) \right]$, where $\boldsymbol{A}$ is the vector potential associated with effective electric field $\boldsymbol{E}^{\mathrm{FM}}$ in FMI. The third order perturbation of hopping yields to the following interfacial ME interactions:

$$\begin{aligned} H_{\mathrm{ME}} &= \xi_{\mathrm{ME}} [\hbar (\dot{\boldsymbol{m}}_i + \dot{\boldsymbol{m}}_j) \cdot (\boldsymbol{m}_i \times \boldsymbol{m}_j) \\ &\quad + 2(\boldsymbol{m}_i \cdot \boldsymbol{m}_j + 1) \delta V_{ij}], \end{aligned} \tag{5}$$

where

$$\xi_{\mathrm{ME}} = \frac{t^2}{(U')^3} e \boldsymbol{E}^{\mathrm{FM}} \cdot (\boldsymbol{r}_j - \boldsymbol{r}_i) \tag{6}$$

is the dimensionless coupling strength and

$$\delta V_{ij} = \frac{1}{2(S+1/2)} \left(\boldsymbol{m}_j \cdot \frac{\partial}{\partial \boldsymbol{m}_j} - \boldsymbol{m}_i \cdot \frac{\partial}{\partial \boldsymbol{m}_i} \right) H_{\mathrm{S}}$$

describes the spin energy change due the electron hopping from site i to site j. H_{S} is related to the spin interactions that do not originate from exchange processes.

The low dimensionality of the FM film would lead to a uniaxial magnetic anisotropy, $H_{\mathrm{S}} = -K_{\mathrm{u}} \boldsymbol{m}_z^2$. The anisotropy K_{u} depends on the FM film thickness d_{FM}, $K_{\mathrm{u}} = \left(K_{\mathrm{s}}/d_{\mathrm{FM}} - \mu_0 M_{\mathrm{s}}^2/2 \right)$, where K_{s} describes the surface anisotropy contributions, which are significant for ultrathin film with magnetization aligned normal to the surface,

whereas, $\mu_0 M_s^2/2$ denotes the demagnetization field that is equivalent to an easy in-plane contribution. Following the above ME discussion, an effective energy density in the continuous limit to the lowest order of $\boldsymbol{m}$ can be written as

$$\mathcal{H} = J_{\rm ex}(\boldsymbol{\nabla}\cdot\boldsymbol{m})^2 - K_{\rm u}\boldsymbol{m}_z^2 + \mathcal{H}_{\rm ME}, \tag{7}$$

where the exchange constant $J_{\rm ex} = t^2/2U'$. $\mathcal{H}_{\rm ME}$ represents the effective ME interaction,

$$\mathcal{H}_{\rm ME} = \frac{4K_{\rm u}\xi_{\rm ME}}{S+1/2}\boldsymbol{m}_z\partial_i\boldsymbol{m}_z + 2\hbar\xi_{\rm ME}\left[\boldsymbol{m}\cdot(\partial_i\boldsymbol{m}\times\partial_l\boldsymbol{m})\right], \tag{8}$$

in which the first and the second terms describe the static and dynamic ME contributions, respectively.

3. Results and discussion

3.1. Static ME effects

By rewriting the magnetization $\boldsymbol{m}(z) = [\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta]$ in the spherical coordinates, the minimum of $\mathcal{H}$ yields the Néel wall-like magnetization configuration. With further considering the Dzyaloshinskii–Moriya interaction stemming from the parity breaking of the lattice at the interfaces, an exponential spiral magnetization (analogous to that in FE/FM metal interface[15]) and even magnetic skyrmions or bubbles, can develop instead,

$$\tan\theta(z) = C\,{\rm e}^{(z-z_0)/\lambda}, \tag{9}$$

where z_0 is the initial position and $1/\lambda = \frac{2K_{\rm u}\xi_{\rm ME}}{S+1/2}/J_{\rm ex}$ with λ being the coherence length of the ME coupling. It should be pointed out that an initial ferromagnetic configuration is considered in the derivation of the above interfacial magnetization. Note that the sign of λ depends on the anisotropy $K_{\rm u}$ and the external electric field; $\mathrm{sign}(\lambda) = \mathrm{sign}(K_{\rm u})\times\langle\hat{\boldsymbol{E}}\cdot\hat{\boldsymbol{e}}_z\rangle$. Along with the boundary conditions determined by the magnetic anisotropy: (i) $\theta(+\infty) = 0$ for out-of-plane magnetization with $K_{\rm u} > 0$ or (ii) $\theta(+\infty) = \pi/2$ for in-plane magnetization with $K_{\rm u} < 0$, one can find that the interfacial nonlinear magnetization can be triggered by only and if only an outward external electric field from the FMI interface, *i.e.*, $\langle\hat{\boldsymbol{E}}\cdot\hat{\boldsymbol{e}}_z\rangle = -1$. This selection rule of the interfacial ME coupling holds promising ways for the electric field-control of domain-wall excitations. On the other hand, following the arguments of ferroelectricity of spin origin,[15,46,47] an inhomogeneous charge distribution can exist due to nonlinear magnetic moments at the interface.[15] As for the typical values: $K_{\rm u} \approx 10$ meV, $U' \approx 1$ eV, $J_{\rm ex} \approx 10$ meV, $\boldsymbol{P}_{\rm s} \approx 10$ mC/cm^2, $\boldsymbol{E}_{\rm c} \approx 10$ kV/cm, and the lattice constant of FMI $a \approx 5$ Å, we obtain a weak (~ 0.1 meV) but long-range ($\lambda \approx 100$ nm) static interfacial ME interaction, which is most pronounced in the vicinity of FE/FMI interface acting with the ME coherence length λ of the order of submicrometers.

3.2. Dynamic ME effects

Without losing of generality, we assume an out-of-plane magnetization, which can be excited by applied (transversal) rf fields $\tilde{\boldsymbol{b}}(t) = [\tilde{b}_x, 0, 0]\,{\rm e}^{{\rm i}\omega t}$,

$$\frac{\partial\boldsymbol{m}}{\partial t} = -\gamma(\boldsymbol{m}\times\tilde{\boldsymbol{b}}), \tag{10}$$

where γ is the gyro-magnetic ratio defined as $\gamma = g\mu_{\rm B}/\hbar$ with the G-factor g and the Bohr magneton $\mu_{\rm B}$. Introducing the above Landau–Lifshitz equation into the ME interaction, Eq. (8), a linear dynamic ME coupling in the lowest order can be expressed as

$$\mathcal{H}_{\rm ME}^{\rm d} = \alpha_{jx}^{\rm ME}\tilde{\boldsymbol{e}}_j\tilde{\boldsymbol{h}}_x, \tag{11}$$

where $\tilde{\boldsymbol{h}}_i = \tilde{\boldsymbol{b}}_i/\mu_{\rm FM}$ is the driving rf magnetic field and $\mu_{\rm FM}$ is the magnetic permeability of FMI. $\tilde{\boldsymbol{e}}_j$ ($j = y, z$) is the rf electric field. According to Eq. (4), the most pronounced dynamic ME coupling occurs if as the rf electric field normal to the interface, *i.e.*, $\tilde{\boldsymbol{e}}_z^{\rm FM} = (\varepsilon_{\rm FE}/\varepsilon_{\rm FM})\tilde{\boldsymbol{e}}_z$, then, the coupling constant reads

$$\alpha_{zx}^{\rm ME} = \frac{4}{a^2}\frac{eJ_{\rm ex}g\mu_{\rm B}\mu_{\rm FM}}{\lambda(U')^2}\frac{\varepsilon_{\rm FE}}{\varepsilon_{\rm FM}}. \tag{12}$$

It should be pointed out that such linear ME effects can be enhanced further in the resonance regime through the magnetic permeability. For typical vaule $\mu_{\rm FM} \approx 10^3\mu_0$, the coupling is estimated to be $\alpha_{zx}^{\rm ME} \approx 10^{-8}$ s/m. The $\alpha_{yx}^{\rm ME}$ does not involve the enhancement ratio $\varepsilon_{\rm FE}/\varepsilon_{\rm FM}$ and it is clearly several orders weaker. As a result, the strongly anisotropic interfacial ME effects appear in the system.

3.3. Transformable magnetic susceptibility

Phenomenologically, the linear ME coupling may result in an additional ME contribution to the magnetization[2,28]

$$\boldsymbol{m}_i = \chi_{ij}^{\rm FM}\tilde{\boldsymbol{h}}_j + \frac{1}{\mu_0}\alpha_{ji}^{\rm ME}\tilde{\boldsymbol{e}}_j, \tag{13}$$

where $\chi_{ij}^{\rm FM}$ is the magnetic susceptibility of FMI, $\chi_{ij}^{\rm FM} = \chi_{\rm m}^{\rm FM} = \mu_{\rm FM}/\mu_0 - 1 \approx \mu_{\rm r}^{\rm FM}$ for isotropic materials. The rf electric fields satisfy the Maxwell–Faraday equation, $\boldsymbol{\nabla}\times\tilde{\boldsymbol{e}} = -\partial\tilde{\boldsymbol{b}}/\partial t$. Taking the propagation of microwave as the $\hat{\boldsymbol{k}}$ direction and $\hat{\boldsymbol{k}}\|(\tilde{\boldsymbol{e}}\times\tilde{\boldsymbol{b}})$, one gets

$$\tilde{\boldsymbol{e}}_j = \frac{\omega}{k}(\hat{\boldsymbol{k}}\times\tilde{\boldsymbol{b}})_j = \frac{c}{\tilde{n}}\mu_{\rm FM}\varepsilon^{jkx}\tilde{\boldsymbol{h}}_x, \tag{14}$$

where c is the speed of light in vacuum, $\tilde{n}$ is the complex refractive index in FMI, and ε^{ijk} is an antisymmetric tensor. By introducing $\tilde{\boldsymbol{e}}_j$ into Eq. (13), one obtains

$$\boldsymbol{m}_x \approx \chi_{\rm m}^{\rm FM}\tilde{\boldsymbol{h}}_x\left[1 + \varepsilon^{jkx}\frac{\alpha_{jx}^{\rm ME}c}{\tilde{n}}\right]. \tag{15}$$

Assuming that $\chi_{\rm m}^{\rm FM}=\chi'_{\rm m}+{\rm i}\chi''_{\rm m}$ and $(1+\varepsilon^{jkx}\alpha_{jx}^{\rm ME}c/\tilde{n})=\beta\,{\rm e}^{{\rm i}\varphi}$, the effective magnetic susceptibility, $\tilde{\chi}_{\rm FM}^{xx}=m_x/\tilde{h}_x$ reads then

$$\Re\tilde{\chi}_{\rm FM}^{xx}=\beta\left[\chi'_{\rm m}\cos\varphi-\chi''_{\rm m}\sin\varphi\right], \tag{16}$$

$$\Im\tilde{\chi}_{\rm FM}^{xx}=\beta\left[\chi'_{\rm m}\sin\varphi+\chi''_{\rm m}\cos\varphi\right], \tag{17}$$

which can be measured by a very sensitive technique, for instance, ferromagnetic resonance (FMR) spectroscopy. Due to the linear ME interaction, the magnetization dynamics is simultaneously influenced by the ac magnetic field $\tilde{\boldsymbol{h}}$ and the ac electric field $\tilde{\boldsymbol{e}}$.[49] The FMR spectrum, thus, is not of a pure absorptive Lorentz mode but an interference mode between the in-phase dispersive ($\chi'_{\rm m}$) and out-of-phase absorptive ($\chi''_{\rm m}$) modes. Furthermore, the mixture phase φ is angular-dependent and electric field-dependent as well, which offers electrical tuning of transformable magnetic susceptibility. More importantly, possible negative magnetic susceptibility could be obtained as $\varphi\approx\pi$. Our recent direct permeability measurements confirmed these dynamic ME effects,[50] in which the linear ME coupling is around $\sim10^{-7}$ s/m in $Co/BaTiO_3$[51] and CoZr/PMN-PT films.[49]

3.4. Negative refraction

In order to explore the effects of the FE/FM interface on the refractive index $\tilde{n}$, the light propagation in the FE/FM heterostructures was investigated. Considering the bound current near the FE/FMI interface, $\boldsymbol{J}=\boldsymbol{\nabla}\times\boldsymbol{M}+\partial\boldsymbol{P}/\partial t$ can be rewritten in the linear response as

$$\boldsymbol{J}_i(\boldsymbol{k},\omega)=K_{ij}(\boldsymbol{k},\omega)\boldsymbol{A}_j(\boldsymbol{k},\omega), \tag{18}$$

where $\boldsymbol{A}(\boldsymbol{k},\omega)$ is the Fourier transformation of the electromagnetic vector potential with the magnetic flux density field $\tilde{\boldsymbol{b}}=\boldsymbol{\nabla}\times\boldsymbol{A}$ and electric field $\tilde{\boldsymbol{e}}=-{\rm d}\boldsymbol{A}/{\rm d}t$. In presence of linear ME effects ($cf.$ Eq. (13), there are three different contributions to the bound current: (i) the magnetization current

$$\boldsymbol{J}_{\rm m}=\boldsymbol{\nabla}\times\boldsymbol{M}=\frac{\chi_{\rm m}^{\rm FM}}{\mu_{\rm FM}}(\delta_{ij}\boldsymbol{k}^2-\boldsymbol{k}_i\boldsymbol{k}_j)\boldsymbol{A}_j,$$

(ii) the polarization current $\boldsymbol{J}_{\rm e}={\rm d}\boldsymbol{P}/{\rm d}t=\varepsilon_0\chi_{\rm e}^{\rm FM}\omega^2\boldsymbol{A}_i$ with $\chi_{\rm e}^{\rm FM}$ being the electric susceptibility of FMI, and (iii) the magnetoelectric current

$$\boldsymbol{J}_i^{\rm ME}=\varepsilon^{i\mu\nu}\partial_\mu m_\nu=\frac{\alpha_{j\nu}^{\rm ME}}{\mu_0}\omega\varepsilon^{i\mu\nu}\boldsymbol{k}_\mu\boldsymbol{A}_j.$$

Assuming the propagation of light in the y direction, $i.e.$, $\hat{\boldsymbol{k}}\|\hat{\boldsymbol{e}}_y$, the bound current then reads

$$\boldsymbol{J}_a=\left[\frac{\chi_{\rm m}^{\rm FM}}{\mu_{\rm FM}}\boldsymbol{k}_y^2+\varepsilon_0\chi_{\rm e}^{\rm FM}\omega^2-\frac{\alpha_{zx}^{\rm ME}}{\mu_0}\omega\boldsymbol{k}_y\right]\boldsymbol{A}_a \quad \text{with} \quad a=z,x. \tag{19}$$

The Maxwell equation

$$\left[\frac{1}{c^2}\frac{\partial^2}{\partial t^2}-\boldsymbol{\nabla}^2\right]\boldsymbol{A}=\mu_0\boldsymbol{J}$$

in the Lorentz gauge yields the following equation for the refractive index $\tilde{n}$:

$$\tilde{n}^2=\mu_{\rm r}^{\rm FM}\varepsilon_{\rm r}^{\rm FM}-\alpha_{zx}^{\rm ME}c\mu_{\rm r}^{\rm FM}\tilde{n}. \tag{20}$$

For the case of $\hat{\boldsymbol{k}}\|\hat{\boldsymbol{e}}_z$, similar equation is obtained but with the replacement of $\alpha_{zx}^{\rm ME}$ by $\alpha_{xy}^{\rm ME}$. It is obvious that we get a standard refractive index $\tilde{n}=\sqrt{\mu_{\rm r}^{\rm FM}\varepsilon_{\rm r}^{\rm FM}}$ in the absence of any ME coupling. Considering ME interactions at the FE/FM interface, a large modification in the refraction index is obtained. For $c\alpha_{ij}^{\rm ME}\approx1$ and the limit of $|\mu_{\rm r}^{\rm FM}|\gg|\varepsilon_{\rm r}^{\rm FM}|$, equation (20) gives two approximate solutions,

$$\tilde{n}_1\approx-\mu_{\rm r}^{\rm FM},\ \ \tilde{n}_2\approx\varepsilon_{\rm r}^{\rm FM}. \tag{21}$$

Note that $|\tilde{n}|=|c/v_{\rm p}|$ (with $v_{\rm p}$ being the phase velocity of light in materials), thus we might have negative refraction ${\rm Re}(\tilde{n})\approx-{\rm Re}(\mu_{\rm r}^{\rm FM})$ in natural nanoscale FE/FM heterostructures. Such negative refraction can exist as well in natural ferromagnetic metals such as nickel, iron, or cobalt at a frequency close to the ferromagnetic resonance if the strength of the mode is large enough.[52]

4. Conclusion

In summary, we have shown that magnetoelectric interactions may develop at the FE/FMI interface due to the nonlocal hopping between multi-orbitals. The large mismatch in dielectric constant of FE and FMI was found to give rise to giant anisotropic electric field effects on magnetization. The interfacial linear dynamic magnetoelectric coupling together with the transformable magnetic susceptibility and the negative refractive index were successfully described. It should be emphasized that the proposed dynamic ME effects are intrinsic phenomena occurring in multiferroic systems with strong linear magnetoelectric interaction and small enough electric permittivity. The predicted properties are of a huge importance in designing in a new class of optical negative-index metamaterials and the obtained results provide new perspectives for construction of the next-generation electronic components based on nanoscale composite multiferroics.

References

[1] Prellier W, Singh M P and Murugavel P 2005 *J. Phys.: Condens. Matter* **17** R803
[2] Eerenstein W, Mathur N D and Scott J F 2006 *Nature* **442** 759
[3] Cheong S W and Mostovoy M 2007 *Nat. Mater.* **6** 13
[4] Spaldin N A, Cheong S W and Ramesh R 2010 *Phys. Today* **10** 38
[5] Vaz C A F 2012 *J. Phys.: Condens. Matter* **24** 333201
[6] Nagaosa N and Tokura Y 2012 *Phys. Scr.* **T146** 014020
[7] Nozaki T, Shiota Y, Miwa S, Murakami S, Bonell F, Ishibashi S, Kubota H, Yakushiji K, Saruya T, Fukushima A, Yuasa S, Shinjo T and Suzuki Y 2012 *Nat. Phys.* **8** 491
[8] Parkin S and Yang S H 2015 *Nat. Nanotechnol.* **10** 209

[9] Kimura T 2012 *Ann. Rev. Condens. Matter Phys.* **3** 93
[10] Koval V, Skorvanek I, Durisin J, Viola G, Kovalcikova A, Svec P, Saksl K and Yan H 2017 *J. Mater. Chem. C* **5** 2669
[11] Vaz C A F, Hoffman J, Ahn C H and Ramesh R 2010 *Adv. Mater.* **22** 2900
[12] Jia C L, Onoda S, Nagaosa N and Han J H 2007 *Phys. Rev. B* **76** 144424
[13] Tokura Y, Seki S and Nagaosa N 2014 *Rep. Prog. Phys.* **77** 076501
[14] Burton J D and Tsymbal E Y 2012 *Phil. Trans. R. Soc. A* **37** 4840
[15] Jia C L, Wei T L, Jiang C J, Xue D S, Sukhov A and Berakdar J 2014 *Phys. Rev. B* **90** 054423
[16] Li Y J, Chen M, Berakdar J and Jia C L 2017 *Phys. Rev. B* **96** 054444
[17] Zhou C, Shen L, Liu M, Gao C X, Jia C L, Jiang C J and Xue D S 2018 *Adv. Funct. Mater.* **28** 1707027
[18] Cui B, Song C, Wang G, Yan Y, Peng J, Miao J, Mao H, Li F, Chen C, Zeng F and Pan F 2014 *Adv. Funct. Mater.* **24** 723
[19] Cui B, Song C, Gehring G A, Li F, Wang G, Chen C, Peng J, Mao H, Zeng F and Pan F 2015 *Adv. Funct. Mater.* **25** 864
[20] Cui B, Song C, Mao H, Yan Y, Li F, Gao S, Peng J, Zeng F and Pan F 2016 *Adv. Mater.* **26** 753
[21] Duan C G, Jaswal S S and Tsymbal E Y 2007 *Phys. Rev. Lett.* **318** 1114
[22] Chakhalian J, Freeland J W, Habermeier H U, Cristiani G, Khaliullin G, van Veenendaal M and Keimer B 2006 *Science* **97** 047201
[23] Benckiser E, Haverkort M W, Brück S, Goering E, Macke S, Frañó A, Yang X, Andersen O K, Cristiani G, Habermeier H U, Boris A V, Zegkinoglou I, Wochner P, Kim H J, Hinkov V and Keimer B 2011 *Nat. Mater.* **10** 189
[24] Cui B, Song C, Mao H, Wu H, Li F, Peng J, Wang G, Zeng F and Pan F 2015 *Adv. Funct. Mater.* **27** 6651
[25] Nagai T, Nagao M, Kurashima K, Asaka T, Zhang W and Kimoto K 2012 *Appl. Phys. Lett.* **101** 162401
[26] Moreo A, Yunoki S and Dagotto E 1999 *Science* **283** 2034
[27] Dagotto E, Hotta T and Moreo A 2001 *Phys. Rep.* **344** 1
[28] Molegraaf H J A, Hoffman J, Vaz C A F, Gariglio S, Van der Marel D, Ahn C H and Triscone J M 2009 *Adv. Mater.* **21** 3470
[29] Eerenstein W, Wiora M, Prieto J L, Scott J F and Mathur N D 2007 *Nat. Mater.* **6** 348
[30] Thiele C, Dörr K, Bilani O, Rödel J and Schultz L 2007 *Phys. Rev. B* **75** 054408
[31] Vaz C A F, Hoffman J, Segal Y, Reiner J W, Grober R D, Zhang Z, Ahn C H and Walker F J 2010 *Phys. Rev. Lett.* **104** 127202
[32] Vaz C A F, Segal Y, Hoffman J, Grober R D, Walker F J and Ahn C H 2010 *Appl. Phys. Lett.* **75** 054408
[33] Ivanshin V A, Deisenhofer J, Krug von Nidda H A, Loidl A, Mukhin A A, Balbashov A M and Eremin M V 2000 *Phys. Rev. B* **97** 042506
[34] Ivannikov D, Biberacher M, Krug von Nidda H A, Pimenov A, Loidl A, Mukhin A A and Balbashov A M 2002 *Phys. Rev. B* **65** 214422
[35] Yunoki S, Moreo A and Dagotto E 1998 *Phys. Rev. Lett.* **81** 5612
[36] Kubo K, Edwards D M, Green A C M, Momoi T, and Sakamoto H 1998 arXiv: cond-mat/9811286 [cond-mat.str-el]
[37] Motome Y, Nakano H and Imada M 1999 *Mater. Sci. Eng. B* **63** 58
[38] Mostovoy M, Nomura K and Nagaosa N 2011 *Phys. Rev. Lett.* **106** 047204
[39] Jackson J D 1998 *Classical Electrodynamics*, 3nd edn. (New York: Wiley) p. 18
[40] Zener C 1951 *Phys. Rev.* **82** 403
[41] Anderson P W and Hasegawa H 1955 *Phys. Rev.* **100** 675
[42] de Gennes P G 1960 *Phys. Rev.* **118** 141
[43] Auerbach A 1994 *Interacting Electrons and Quantum Magnetism* (Berlin: Springer-Verlag) pp. 25–28
[44] Graf M and Vogl P 1995 *Phys. Rev. B* **51** 4940
[45] Ismail-Beigi S, Chang E K and Louie S G 2001 *Phys. Rev. Lett.* **87** 087402
[46] Mostovoy M 2006 *Phys. Rev. Lett.* **96** 067601
[47] Betouras J J, Giovannetti G and Van den Brink J 2007 *Phys. Rev. Lett.* **98** 257602
[48] Engelbrecht S, Shuvaev A M, Luo Y, Moshnyaga V and Pimenov A 2011 *Europhys. Lett.* **95** 37005
[49] Jia C L, Wang F L, Jiang C J, Berakder J and Xue D S 2015 *Sci. Rep.* **5** 11111
[50] Jiang C J, Jia C L, Wang F L, Zhou C and Xue D S 2018 *Phys. Rev. B* **97** 060408
[51] Jedrecy N, von Bardeleben H J, Badjeck V, Demaille D, Stanescu D, Magnan H and Barbier A 2013 *Phys. Rev. B* **88** 121409
[52] Engelbrecht S, Shuvaev A M, Luo Y, Moshnyaga V and Pimenov A 2013 *Europhys. Lett.* **95** 37005