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## Landau–Zener tunneling in multiferroic composites

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## PAPER

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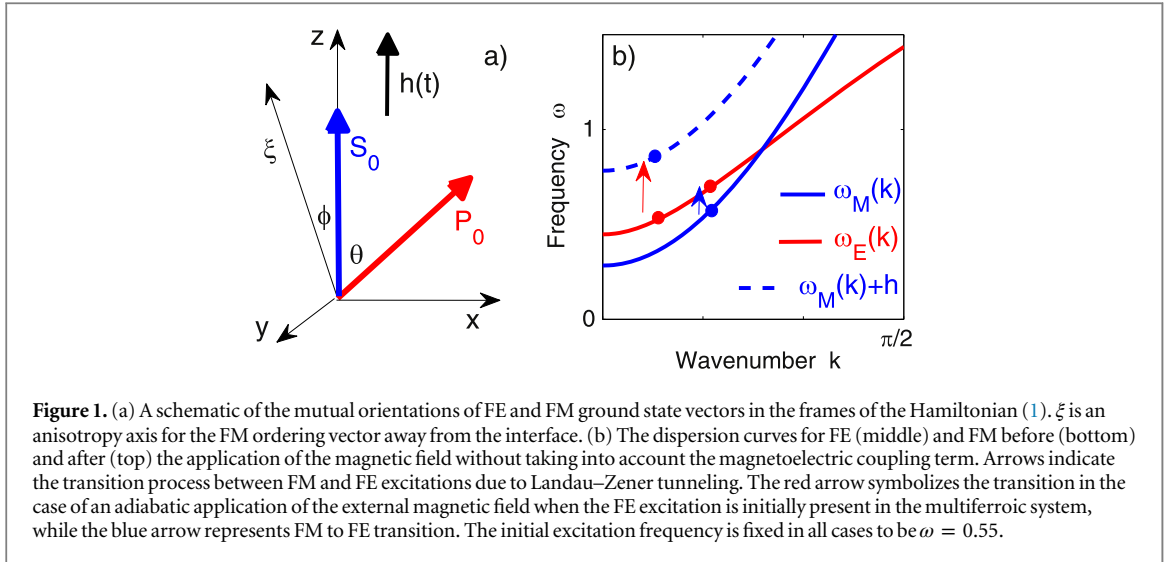
**Abstract**

For a coupled ferroelectric/ferromagnetic composite we demonstrate theoretically a mechanism for ferroelectric–ferromagnetic excitation conversion. The switching occurs upon sweeping an external homogeneous magnetic field that modifies the spectrum. We show analytically and by direct numerical simulations that the excitation transfer proceeds via Landau–Zener tunneling: in the case of an abrupt application of the magnetic field (a diabatic regime) there is no transition of excitation energy between the electric and magnetic systems, while increasing the magnetic field slowly (an adiabatic process) an almost complete excitation transfer takes place.

**Introduction**

Originally Landau–Zener tunneling was theoretically predicted [1, 2] for purely quantum objects. The phenomenon is realized when, upon a parametric change in the Hamiltonian, a crossing of the unperturbed atomic levels is expected to occur. The degeneracy is lifted by a perturbation and the transition probability between the states depends not only on their coupling, but also on the speed of the parametric change of the energy level spacing. Later, Landau–Zener tunneling was generalized from microscopic quantum systems (such as electron motion in semiconductor superlattices [3], level transitions in Rydberg [4] and hydrogen [5] atoms and tunneling in double quantum dots [6]) to macroscopic wave processes in different branches of physics; for instance, matter wave mini-band transitions in Bose–Einstein condensates [7, 8], light tunneling in optical waveguide arrays [9–12] and two-dimensional optical lattices [13], macroscopic tunneling in classical optical systems [14], and even ultrasonic transitions in water cavity superlattices [15] and vibrational transport in harmonic mechanical oscillator systems [16]. Further important aspects are the influence of dissipation [17, 18] or temperature induced fluctuations on the Landau–Zener transitions [19] as well as the nonlinear [20–22] and multiple [23] versions of asymmetric Landau–Zener tunneling.

An essential ingredient of all the aforementioned cases is that a parametric change modifies the energy level spacing. In the present paper this parameter is an external magnetic field applied to a heterostructure consisting of a ferromagnet (FM) coupled to adjacent ferroelectrics (FE), thus forming composite multiferroics. Examples are Fe or Co layers deposited on the prototypical FE BaTiO<sub>3</sub>. Such systems were recently fabricated and were shown to exhibit a magnetoelectric response [24, 25] (see also [26]), i.e. a FM (FE) response to an applied electric (magnetic) perturbation. Here we demonstrate, using a theoretical model, that an external field may result in Landau–Zener phenomena leading to electric–magnetic (dipolar-spin) polarization excitation transfer. Thereby, the presence of interfacial weak magnetoelectric coupling between the FE and FM parts is essential. Tunneling transitions occur when, with increasing magnetic field, the frequency of magnetic excitations (belonging to the magnetic branch of the multiferroic) with fixed wavenumber shifts upward to cross the electric excitation dispersion branch at the same wavenumber. Sweeping the magnetic field adiabatically, one observes a complete transition of the initial electric (or magnetic) excitations to magnetic (or electric) ones, while in the diabatic case almost no tunneling of electric or magnetic excitations takes place. This finding suggests an externally controlled and effective method for conversion of electric/magnetic pulses in FE/FM heterostructures.



**Figure 1.** (a) A schematic of the mutual orientations of FE and FM ground state vectors in the frames of the Hamiltonian (1).  $\xi$  is an anisotropy axis for the FM ordering vector away from the interface. (b) The dispersion curves for FE (middle) and FM before (bottom) and after (top) the application of the magnetic field without taking into account the magnetoelectric coupling term. Arrows indicate the transition process between FM and FE excitations due to Landau–Zener tunneling. The red arrow symbolizes the transition in the case of an adiabatic application of the external magnetic field when the FE excitation is initially present in the multiferroic system, while the blue arrow represents FM to FE transition. The initial excitation frequency is fixed in all cases to be  $\omega = 0.55$ .

## Theoretical modeling

For insight into the time dynamics of the correlated electric dipoles and magnetic moments in multiferroics [27–29] we consider a minimal but relevant ladder model consisting of two weakly coupled chains: the first chain is FE, built by one-dimensional interacting dipoles, and the second chain is FM, consisting of classical three-dimensional magnetic moments. In the case of both single [30–32] and two-phase [33–36] multiferroics, both the FE and FM chains are characterized by intrachain nearest neighbor coupling.

For predictions that are experimentally relevant we consider a heterojunction consisting of a well-studied conventional FM such as iron and a prototypical FE such as BaTiO<sub>3</sub>. The theoretical model should reproduce the known behavior of the FE and the FM order parameters in the absence of magnetoelectric coupling. In our study we model the FM part using the well-established classical Heisenberg model and choose iron as the FM. For FE we utilize the Ginzburg–Landau–Devonshire (GLD) potential [37–40]. The bi-harmonic potential employed here corresponds to BaTiO<sub>3</sub> in the tetragonal phase (see [41, 42] and references therein). This system has been experimentally realized, as reported in [25, 38] where an iron film a few atomic layers thick was deposited on a single-crystal BaTiO<sub>3</sub>. The FE/FM coupling may have different origins, here we assume the driving source to be a spin-dependent dynamic screening effect at the FE/FM interface [43, 44]. The strength of the coupling has been evaluated using first principles calculations.

In the frame of this model each one-dimensional electric dipole  $P_n$  is coupled with the nearest spin  $\vec{S}_n$  in the ladder via an interchain weak magnetoelectric coupling. Here we assume the form given in [44] that is linear in the magnetization and polarization. Experiments seem to confirm this type of coupling [24]. We assume the direction of the FE dipoles to be at some arbitrary angle with respect to FM anisotropy axis  $\xi$  depicted in figure 1(a). Magnetoelectric coupling causes rearrangement of magnetic moments in FM. The new ground state ordering direction in FM we identify by the axis  $z$  and  $\phi$ , which is the angle between  $z$  and the anisotropy axis  $\xi$ . Furthermore, we assume that a magnetic field  $\vec{h}(t)$  is applied along this new ordering direction  $z$ . At the same time,  $\theta$  is an angle between the  $z$  axis and the direction of the FE dipoles (see figure 1(a)).  $S_0$  and  $P_0$  represent the equilibrium values of the spins and the dipoles, respectively. We will consider perturbations around the equilibrium. Particularly, let us define the dipolar deviations  $p_n \equiv (P_n - P_0)/P_0$  and the reduced spin variables  $\vec{s}_n \equiv \vec{S}_n/S_0$  and write the model Hamiltonian in the following form:

$$\begin{aligned}
 H &= H_P + H_S + H_{SP}, & H_{SP} &= -\tilde{g} \sum_{n=1}^N p_n s_n^x, \\
 H_P &= \sum_{n=1}^N \frac{\tilde{\alpha}_0}{2} \left( \frac{dp_n}{dt} \right)^2 + \frac{\tilde{\alpha}}{2} p_n^2 + \frac{\tilde{\beta}}{4} p_n^4 + \frac{\tilde{\alpha}_J}{2} (p_{n+1} - p_n)^2, \\
 H_S &= \sum_{n=1}^N \left[ -\tilde{J} \vec{s}_n \vec{s}_{n+1} + \tilde{D}_1 (s_n^x)^2 + \tilde{D}_2 (s_n^y)^2 - \tilde{h}(t) s_n^z \right], \tag{1}
 \end{aligned}$$

where  $H_{SP}$  describes the magnetoelectric coupling (with coupling constant  $\tilde{g}$ ). In general,  $H_{SP}$  may include higher order terms in  $p_n$  and  $s_n^j$ . For instance, in the case of a quadratic coupling  $-\tilde{g}_1 (\vec{P}_n \vec{S}_n)^2$  the coupling constant

attains the form  $\tilde{g}_1 S_0^2 P_0^2 \sin(2\theta)$  ( $\theta$  being the angle between  $P_0$  and  $\vec{S}_0$ , see figure 1(a)).  $H_p$  is the Hamiltonian of the FE part of the multiferroic system, describing  $N$ -interacting FE dipoles [41, 45]. We recall that in the spirit of a coarse-grained treatment the functional structure of (1) is inherent to the symmetry and dimension of the FE and FM order parameters. The microscopic material-specific properties, i.e., whether we are dealing with BaTiO<sub>3</sub>/Fe or another FE/FM composite, are determined by Landau's kinetic coefficients  $\tilde{\alpha}_0, \tilde{\alpha}_J$  (the nearest neighbor coupling constant),  $\tilde{\alpha}$  and  $\tilde{\beta}$  (second and fourth order expansion coefficients of the GLD potential [37, 41] near the equilibrium state  $P_0$ ). The numerical values of these parameters are specified below.  $H_S$  describes the FM chain [46] with  $\tilde{J}$  being the nearest neighbor exchange coupling in the FM part.  $\tilde{D}_1 = \tilde{D}S_0 \cos^2 \phi$  and  $\tilde{D}_2 = \tilde{D}S_0$  are anisotropy constants, where  $\tilde{D}$  is the uniaxial anisotropy constant along axis  $\xi$  (see figure 1(a)).  $\tilde{h}(t)$  is a time-dependent homogeneous magnetic field responsible for Landau-Zener tunneling between dipolar and spin excitations.

For clarity, we introduce a dimensionless, scaled time as  $t \rightarrow \omega_0 t$  ( $\omega_0 = \sqrt{\tilde{\alpha}_J/\tilde{\alpha}_0} \sim 10^{12}$  Hz) and measure all the coefficients  $\tilde{g}, \tilde{\alpha}, \tilde{\beta}, \tilde{J}, \tilde{D}_1, \tilde{D}_2$  and the external magnetic field  $\tilde{h}(t)$  in units of  $\omega_0$  and omit the tilde superscript (for instance we introduce rescaled constants  $g = \tilde{g}/\omega_0$ , etc). The equations governing the multiferroic time evolution read

$$\begin{aligned} \frac{\partial s_n^x}{\partial t} &= -J \left[ s_n^y (s_{n-1}^z + s_{n+1}^z) - s_n^z (s_{n-1}^y + s_{n+1}^y) \right] \\ &\quad - h(t) s_n^y - 2D_2 s_n^y s_n^z, \\ \frac{\partial s_n^y}{\partial t} &= J \left[ s_n^x (s_{n-1}^z + s_{n+1}^z) - s_n^z (s_{n-1}^x + s_{n+1}^x) \right] \\ &\quad + h(t) s_n^x + 2D_1 s_n^x s_n^z - g p_n s_n^z, \\ \frac{d^2 p_n}{dt^2} &= -\alpha p_n - \beta p_n^3 + (p_{n-1} - 2p_n + p_{n+1}) + g s_n^x. \end{aligned} \quad (2)$$

We are concerned with small perturbations around the equilibrium, i.e., the variables  $s_n^x, s_n^y$  and  $p_n$  are much smaller than unity. Therefore,  $s_n^z$  could be computed using the approximate equality

$$s_n^z = 1 - (s_n^x)^2/2 - (s_n^y)^2/2.$$

## Analytical considerations

For an analytical consideration of the processes taking place upon the application of a time-dependent magnetic field, we neglect in (2) all nonlinear terms (setting  $s_n^z = 1$  and omitting the term proportional to  $p_n^3$ ). The resulting equations read

$$\begin{aligned} \frac{\partial s_n^x}{\partial t} &= J (s_{n-1}^y + s_{n+1}^y - 2s_n^y) - 2D_2 s_n^y - h(t) s_n^y, \\ \frac{\partial s_n^y}{\partial t} &= -J (s_{n-1}^x + s_{n+1}^x - 2s_n^x) - 2D_1 s_n^x + h(t) s_n^x - g p_n, \\ \frac{d^2 p_n}{dt^2} &= (p_{n+1} + p_{n-1} - 2p_n) - \alpha p_n + g s_n^x. \end{aligned} \quad (3)$$

We will solve these coupled equations perturbatively by seeking a solution as an expansion in the relative time-smoothness, i.e.

$$\begin{aligned} \begin{pmatrix} s_n^x \\ s_n^y \end{pmatrix} &= \left[ \psi(t) \mathbf{R} + \mathcal{O}_1(\varepsilon) \right] e^{i(kn - \omega t)} + \text{c. c.} \\ p_n &= \left[ \varphi(t) + \mathcal{O}_2(\varepsilon) \right] e^{i(kn - \omega t)} + \text{c. c.} \end{aligned} \quad (4)$$

where the functions  $\psi(t)$  and  $\varphi(t)$  are slowly varying in time (i.e.  $\dot{\psi} \equiv d\psi/dt \ll \omega\psi$ ) and the small parameter  $\varepsilon$  quantifies the relative time-smoothness of  $\psi(t)$  and  $\varphi(t)$ , i.e.  $\varepsilon \sim \dot{\psi}/(\omega\psi)$ ;  $\mathbf{R}$  is a two component column vector with complex components. Let us assume that the terms proportional to  $h(t)$  and  $g$  are of the order of the small parameter  $\varepsilon$  and could be neglected in a first approximation. Then the equations in leading order are decoupled into magnetic and FE parts. Inserting (4) into (3), the first two equations are reduced to the matrix equality

$$\hat{\mathbf{W}} * \mathbf{R} = 0, \quad \hat{\mathbf{W}} = \begin{pmatrix} -i\omega & 4J \sin^2 \frac{k}{2} + 2D_2 \\ -4J \sin^2 \frac{k}{2} - 2D_1 & -i\omega \end{pmatrix}.$$

Then the solvability condition  $\text{Det}(\hat{\mathbf{W}}) = 0$  yields the dispersion relation

$$\omega = \omega_M(p) = 2\sqrt{\left(2J \sin^2 \frac{k}{2} + D_2\right)\left(2J \sin^2 \frac{k}{2} + D_1\right)} \quad (5)$$

for magnetic spins without taking into account magnetoelectric coupling and applied external magnetic field. On the other hand, we can write the expression for the column vector  $\mathbf{R} = (1, ir)$ , where  $r = 2\left(2J \sin^2 \frac{k}{2} + D_1\right)/\omega_M$ , and introduce a row vector  $\mathbf{L}$  satisfying the matrix relation  $\mathbf{L} * \hat{\mathbf{W}} = 0$  and thus obtaining the following components:  $\mathbf{L} = (1, -i\ell)$  where  $\ell = 2\left(2J \sin^2 \frac{k}{2} + D_2\right)/\omega_M$ .

With all these results, derived from the leading perturbative limit, being inserted in (3) and summing the product of the first two equations on the respective components of row vector  $\mathbf{L}$  and noting that  $r\ell = 1$ , we find the next approximation

$$\begin{aligned} 2\frac{\partial\Psi}{\partial t} &= i\left[2(\omega - \omega_M) - (\ell + r)h(t)\right]\Psi + i\ell g\varphi, \\ -2i\omega\frac{\partial\varphi}{\partial t} &= (\omega^2 - \omega_E^2)\varphi + g\Psi, \end{aligned} \quad (6)$$

where

$$\omega_E = \sqrt{\alpha + 4 \sin^2 \frac{k}{2}} \quad (7)$$

represents the frequency of the electric dipoles while neglecting the magnetoelectric coupling. Introducing a new variable  $\Phi = \varphi\sqrt{\omega\ell}$ , shifting the phases of  $\Phi$  and  $\Psi$  and rescaling the time, we retrieve a Landau–Zener problem from (6) in the standard form, i.e.,

$$i\frac{\partial\Psi}{\partial t} = -\nu t\Psi + \mu\Phi, \quad i\frac{\partial\Phi}{\partial t} = \nu t\Phi + \mu\Psi, \quad (8)$$

where we have assumed a linear time dependence of the external magnetic field  $h(t) = h_0 t$ . The slope  $\nu$  and the coupling  $\mu$  coefficients are defined by the following relations

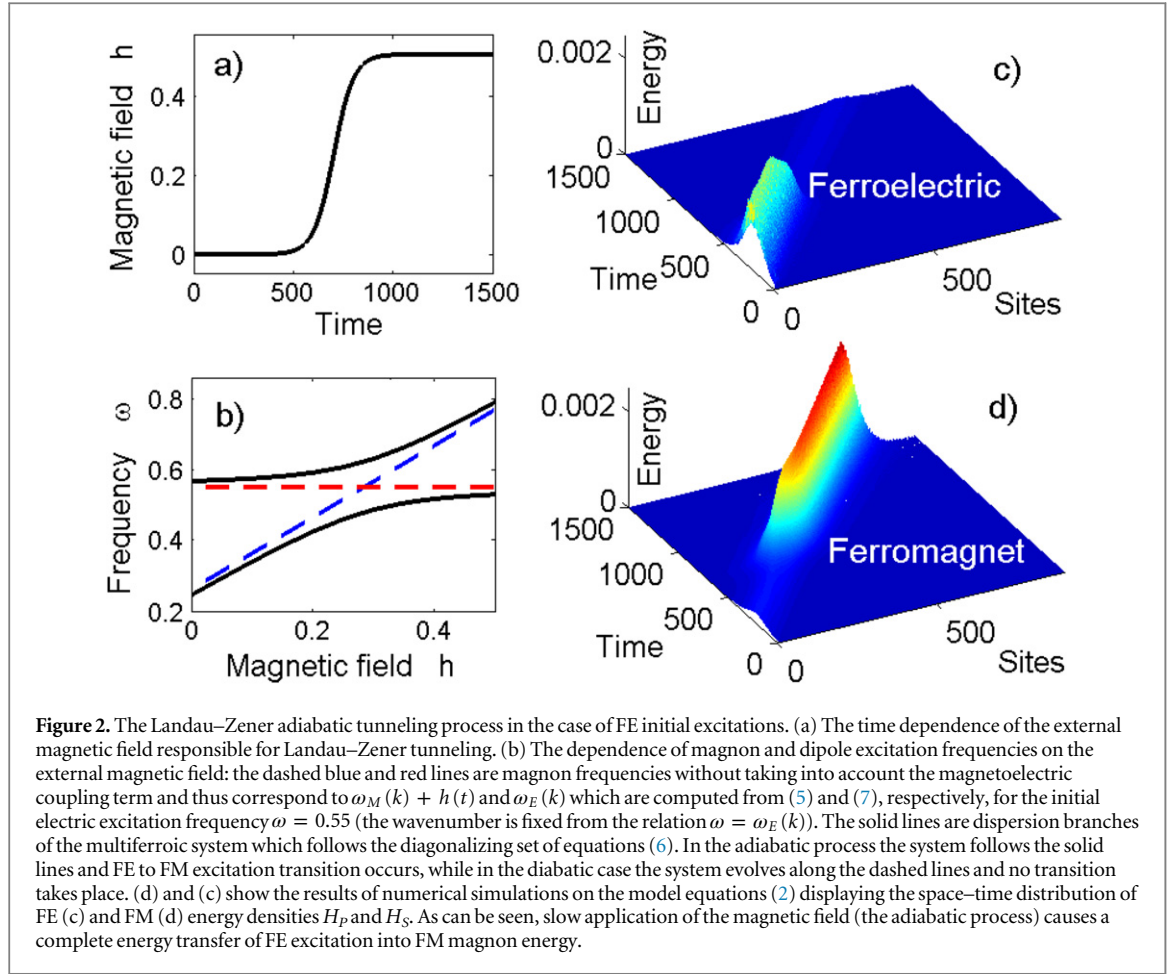
$$\nu = h_0(r + \ell), \quad \mu = g\sqrt{\ell/\omega}, \quad (9)$$

and we infer the well known Landau–Zener formula [1, 2] for the tunneling probability of magnetic excitations to FE excitations and vice versa

$$\kappa = \exp\left[-\frac{\pi\mu^2}{\nu}\right] = \exp\left[-\frac{\pi g^2 \ell}{h_0 \omega (r + \ell)}\right]. \quad (10)$$

The Landau–Zener mechanism operates as follows: according to the equations (8), if at  $t \rightarrow -\infty$  one has  $\Psi = \Psi_0$  and  $\Phi = 0$  then at  $t \rightarrow \infty$  the function  $\Psi$  takes the value  $|\Psi_f|^2 = \kappa |\Psi_0|^2$ , while  $|\Phi_f|^2 = (1 - \kappa) |\Psi_0|^2$ . Similar relations hold if at  $t \rightarrow -\infty$  one starts with  $\Psi = 0$  and  $\Phi = \Phi_0$ . Particularly, at  $t \rightarrow \infty$  we obtain  $|\Psi_f|^2 = (1 - \kappa) |\Phi_0|^2$ ,  $|\Phi_f|^2 = \kappa |\Phi_0|^2$ . The functions  $\Psi$  and  $\Phi$  can easily be related with the electric and magnetic moments by considering the definitions (4),  $|\Psi|^2 = (\langle s_n^\perp \rangle^2 / 2)(1 + r^2)$  (where  $\langle (s_n^\perp)^2 \rangle \equiv \langle (s_n^x)^2 \rangle + \langle (s_n^y)^2 \rangle$ ) and  $|\Phi|^2 = |\phi|^2 \omega \ell = \omega \ell \langle (p_n)^2 \rangle / 2$ . Here  $\langle \dots \rangle$  denotes time averaging. In our numerical simulations we always initially have a mixed state of electric and magnetic excitations and, knowing the initial and final averaged values of the electric and magnetic excitations, after simple algebra one can calculate the tunneling probability coefficient  $\kappa$  according to the following formula:

$$\kappa = \frac{|\Psi_f|^2 - |\Phi_0|^2}{|\Psi_0|^2 - |\Phi_0|^2} = \frac{\langle (s_n^\perp)^2 \rangle_f - \omega \ell (1 + r^2) \langle (p_n)^2 \rangle_0}{\langle (s_n^\perp)^2 \rangle_0 - \omega \ell (1 + r^2) \langle (p_n)^2 \rangle_0}. \quad (11)$$



## Numerical results and analysis

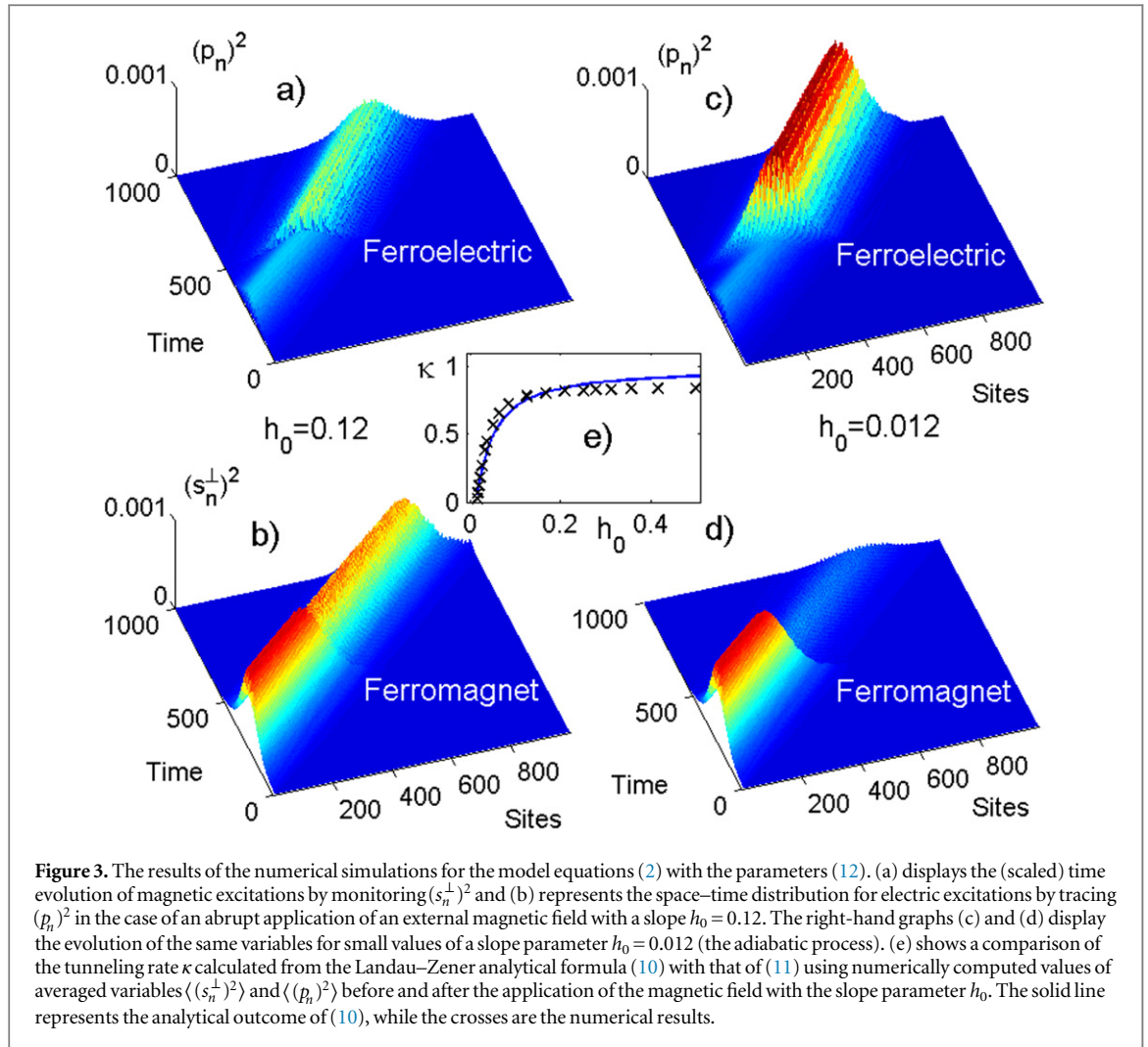
To check the reliability of the analytical predictions and the validity of the assumptions, we conducted full numerical simulations for the evolution equations (2) with the following parameters in reduced dimensionless units:

$$\alpha = 0.2; \beta = 0.1; J = 1; D_1 = 0.1; D_2 = 0.2; g = 0.1. \quad (12)$$

While the model does not incorporate all facets that are of importance in a real experimental set up, these parameter do mimic realistic materials, namely BaTiO<sub>3</sub>/Fe, as follows from *ab initio* calculations [47, 48] and experimental findings [24]. The essential parameters to be entered in (1) are (for more details see [42]): the FE second and fourth order potential coefficients  $\tilde{\alpha}_1/(a_{\text{FE}}^3) = 2.77 \cdot 10^7$  (Vm C<sup>-1</sup>),  $\tilde{\alpha}_2/(a_{\text{FE}}^3) = 1.7 \cdot 10^8$  (Vm<sup>5</sup> C<sup>-3</sup>), the FE coupling coefficient  $\tilde{\alpha}_J/(a_{\text{FE}}^3) = 1.3 \cdot 10^8$  (Vm C<sup>-1</sup>), the equilibrium polarization  $P_0 = 0.265$  (C m<sup>-2</sup>) and the coarse-grained FE cell size  $a_{\text{FE}} = 1$  (nm). The FM exchange interaction strength is  $\tilde{J}_s = 3.15 \cdot 10^{-20}$  (J), the FM anisotropy constant is  $\tilde{D} = 6.75 \cdot 10^{-21}$  (J), and the ME coupling strength is  $\tilde{g}_1 \approx 10^{-21}$  (Vm<sup>2</sup>).

In the numerical simulations, for initially excited electric (or magnetic) degrees of freedom in the multiferroics we apply a time-varying magnetic field and monitor the time evolution of those excitations. First we preferably excite electric dipoles (their dispersion relation is indicated by the red solid line in figure 1(b)) with an oscillation frequency  $\omega = 0.55$  fixing the wavenumber  $k$  from the dispersion relation  $\omega = \omega_E(k)$  where  $\omega_E(k)$  is defined from (7). Then, the magnetic field is gradually applied with the following time dependence:  $h(t) = 0.25[1 - \tanh((t - t_0)/\tau)]$  where  $t_0$  is half of the computation time (see figure 2(a)). By changing  $\tau$  we vary the slope of the magnetic field  $h_0 = 0.25/\tau$  (Landau–Zener tunneling takes place in the vicinity of  $t = t_0$  and thus this definition of slope is a good approximation). This tunneling happens because the magnon frequency (in figure 1(b) the blue solid line describes the magnon dispersion relation in the absence of the external magnetic field) during the application of the magnetic field shifts upward (the blue dashed line in the same graph) and traverses the electric frequency curve. If the variation of the magnetic field is adiabatic (small slope values  $h_0$ ) then the energy of the electric excitations completely transfers to magnons (this case is considered in figures 2(c) and (d), and is indicated by the red line in figure 1(b)). Otherwise, in the diabatic case (large values of





$h_0$ ), the energy remains within the electric excitations. Both the adiabatic and diabatic processes could be schematically explained by inspecting the graph in figure 2(b), where the solid lines display the dispersion branches of the multiferroic derived by the diagonalization of (6). In the case of an adiabatic increase of the magnetic field, the system follows the upper solid curve transferring the energy from electric to magnetic excitations. In the diabatic case of magnetic field applications the system follows the red dashed curve and the energy persists within the electric excitations.

To check how well the Landau–Zener formula (10), (11) describes the energy exchange between the electric and the magnetic degrees of freedom, we have undertaken detailed studies of the tunneling process for various slopes of the external magnetic field, while initially exciting the magnetic moments. In figure 3 we show the numerical outcome of the case when preferably magnetic excitations (again with the same  $\omega = 0.55$  frequency and thus fixed wavenumber  $k$  from the magnon dispersion relation  $\omega = \omega_M(k)$ ) are present in the system. Applying a magnetic field with different slopes  $h_0$  one monitors either an absence of switching of magnetic excitations into electric ones ((a) and (b) of figure 3, which correspond to a large slope value  $h_0 = 0.12$ ), or an almost complete transfer of energy to the electric dipole excitations ((c) and (d) of figure 3 for the small slope value  $h_0 = 0.012$ ). This transition process is schematically indicated by the blue vertical arrow in figure 1(b). Moreover, the process can be identified as an evolution along the lower solid line of figure 2(b) in the case of an adiabatic change of the external magnetic field. In the diabatic case, in contrast, the system follows the blue dashed line of the same graph. A wide range of the slope parameter  $h_0$  has been considered and the tunneling rates have been computed according to (11), the numerical results indicated by the crosses in figure 3(e). These numerical points are in a good agreement with the Landau–Zener analytical formula (10) given by the solid line in the same graph. A discrepancy appears for large slope values when their values are comparable with the excitation frequency. This violates the condition that the magnetic field should vary much slower than the dipole or the spin precession frequency (recall that (8) were derived in just this approximation  $h_0 \ll \omega$ ).

In this paper we do not address dissipation and thermal effects. In principle these could be taken into account by introducing the conventional Landau–Lifshitz–Gilbert damping term in the magnetic part of the

evolution equations (2) as well as the damping terms in the electric part; while stochastic thermal processes could be described by random fluctuating magnetic and electric fields. Then the Landau–Zener transition processes would be modified according to the mechanisms described in [17–19]. Here, we assume that dissipation is negligible on time scales comparable to magnetic/electric signal transition (that is  $\sim 10^{-9}$  s) and do not consider respective terms in the evolution equations. For the case of multiferroic composites for which the electromagnetic coupling coefficient is much weaker than that considered here, the magnetic field variation should be much slower in order to achieve the transition from electric to magnonic excitations or vice versa. Dissipation and fluctuation processes may become relevant.

## Summary

In conclusion, an effective mechanism for an externally controlled energy exchange between electric dipole and magnetic excitations in composite multiferroics has been suggested. The method relies on time varying an applied magnetic field that affects the excitation spectra and may lead to Landau–Zener tunneling phenomena. We considered this mechanism analytically and numerically and find excellent quantitative agreement between the analytical formula for the tunneling rate and the results from direct numerical simulations. The effect could be directly applied for a complete conversion of electric into magnetic signals or vice versa. On the other hand, via the Landau–Zener mechanism one could study electric and magnetic excitation branches of given multiferroic composites and estimate the electromagnetic coupling coefficients. Indeed, the transition probabilities depend strongly on that coefficient. One can measure the coupling coefficient by investigating the transition probabilities while varying diabatically the application of the magnetic field. As a further perspective, this control mechanism could be extended in a straightforward manner to nonlinear excitations, where the asymmetric character [20–22] of the Landau–Zener tunneling process could be realized.

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