A simple model for the magnetoelectric interaction in multiferroics

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Abstract. The (anti)ferromagnetic and ferroelectric transitions in some multiferroic compounds seem to be strongly correlated. Even for systems that do not show spontaneous ferroelectricity such as the LiMPO$_4$ (M = Mn, Fe, Co, Ni) compounds, the coupling between magnetic and electric degrees of freedom is evident experimentally. Here, we present a simple numerical calculation to simulate this coupling that leads to the two transitions. We assume a magnetic sublattice consisting of classical magnetic moments coupled to a separated non-magnetic sublattice consisting of classical electric dipoles. The coupling between them is realized through a phenomenological spin-lattice Hamiltonian, and the solution is obtained using the Monte Carlo technique. In the simplest version, the magnetic system is 2D Ising (anti)ferromagnetic lattice, with nearest neighbors interactions only, and the electric moments are permanent moments, coupled electrically. Within this approximation, the second order magnetic transition induces ferroelectricity in the electric dipoles. We show that these calculations can be extended to other magnetic systems, (x-y model and 3D Heisenberg) and to systems where the electric moments are created by strains, generated via spin-lattice coupling, so the model can be applied to model realistic systems such as the olivines mentioned above.

1. Introduction

Recently, there has been a revived interest in the research of multiferroics due to several new discoveries, and the possibility to use them technologically [1]. The electric and magnetic transitions are not necessarily correlated, but when it occurs - the so called magnetoelectric effect appears - the materials suggest possible use as devices. The LiMPO$_4$ (M = Mn, Fe, Co, Ni) compounds generated great interest in the last times, for they are used as electrodes in batteries, but also because they present very rich physics [1-2]. The whole family is multiferroic (that is, present simultaneously magnetic and electric order), with transitions related one to the other, and their transitions show very special effects. The Ni compound undergoes a first order phase transition from collinear antiferromagnetic ground state to long-range incommensurate structure at $T_N = 20.8$ K and at $T_{IC} = 21.7$ K the incommensurate structure transforms to short range [2]. Inelastic and elastic neutron studies reveal anomalous spin wave in this incommensurate phase, and iron substitution (about 20%) fine tunes the phase transition and the spin-wave dispersion [2]. Many other interesting physical properties of these compounds are in the recent literature. However, just few papers are dedicated to the theoretical interpretation of the magnetoelectric effect [4].
We present here a simple and understandable model for the magnetoelectricity in these and similar compounds. The electromagnetic Hamiltonian is solved for very simple cases, and the solutions present similitude with the reported experiments. The model is extended to more complicated systems, and partially solved in those cases too. These results and comparisons will be published elsewhere.

2. The model

We assume that the magnetoelectric system is a set of magnetic dipoles, coupled via the exchange interaction, in a lattice with a distribution of electric charges, susceptible to change when the magnetic dipoles change their orientations. The change in orientation of the magnetic dipoles modify their environment, via spin-orbit interaction, creating local strains, and creating or orienting a set of electric dipoles in the lattice. We assume that our crystal suffers the strain in such a way that electric dipoles are oriented to a particular direction when the magnetic dipoles relax.

The model Hamiltonian used in this work follows:

\[ H = H_M + H_E + H_{ME} \]  

where \( H_M \) is the magnetic energy, \( H_E \) the electric energy and \( H_{ME} \) the magnetoelectric coupling.

2.1. Approximations

The first approach for a solution of eq. (1) is obtained replacing the first term in the sum of the right side by a square sublattice of Ising magnetic moments, and the electric moments in the second and third terms by random oriented classical electrical dipoles, located in a separated square sublattice. The Ising spins are coupled to their nearest neighbors only, and with periodic boundary conditions. The interaction Hamiltonian allows only nearest neighbors magnetoelectric interaction. Symmetry requires that the magnetic point group of the magnetic moment is one of the 58 Shubnikov groups that allow magnetoelectricity [1]. This forces the magnetic moments to have one electric dipole only as a nearest neighbor.

The numerical solution of the problem was done using the importance sample Monte Carlo method, looking for the minimum in energy for our system.

Thus,

\[ H = -J \sum_{<i,j>} \sigma_i \sigma_j - h \sum_{<i,j>} \sigma_i - \beta \sum_{<i,j>} P_{ix} P_{jx} + \alpha \sum_i P_{ix} \]  

is the approximated Hamiltonian, where \( J \) is the exchange coupling of the Ising magnetic spins \( \sigma \), and \( P \) the electric dipoles. The symbol \( <i,j> \) indicates sums over the nearest neighbors only, in both the electric and magnetic cases. The first and second term constitutes the magnetic energy, where we included the possibility of an applied or external magnetic field \( h \). The third term represents the electric energy, proportional to the orientation of neighbor electric momenta. The interaction term, which represents a spin - lattice Hamiltonian, has been extremely simplified. As the interaction energy would depend of the value of the created dipole (or strain), it is proportional to the projection of the electric dipole in the selected direction; the value of the coupling, contained in the \( \alpha \) parameter, which also contains the value of the magnetic moment. This very simple expression is convenient to allow the values of the coupling to be positive or negative, as the strain can add or take energy of the lattice.
2.2. Ferromagnetic case.

Our first calculation was performed in a 100×100 2D Ising ferromagnetic spins coupled to a separated 100×100 electric dipoles. The electric dipoles were oriented at random, as was the magnetic lattice, to begin with infinite temperature. The temperature was then fixed to a value, and a Monte Carlo program, where the transitions are allowed following the Metropolis technique, is iterated the time necessary to obtain thermal equilibrium of the system. Then, the converged results are used as initial condition for the following temperature. The calculation was performed reducing the temperature in each step.

The complete calculation was performed after a study of convergence of our model. As first step, we did not considered the electric and interaction energies when we looked for the minimum. This means that the model is just a 2D Ising system, moving electric dipoles together, and as expected, the magnetization follows the Ising model as was done before. The exact calculation published by Onsager allows a very good comparison, and the electric dipoles also feel a transition at the same temperature.

Figure 1. (Color online) The results for the ferromagnetic case. a) Magnetization as a function of $T$, for different values of $\alpha$; b) The same for different values of $\beta$; c) The electric polarization as function of the temperature, for different values of $\beta$; d) The complete dependence of $T_c$ as a function on the parameters of the model.

The following step was to study the convergence when the parameters $\alpha$ and $\beta$ in the model are different from zero. It is well known that the Ising model converges slowly near by the transition temperature, due to fluctuations, and the equal value for the energy when the system is oriented in any of the both possible directions. This is easily solved with the addition of the small external field $h$; however, we observed that for particular values of $\alpha$, the convergency is
the slowest. This can be explained by the fact that $\alpha$ appears in the Hamiltonian as an extra external field, in some manner. The second and last terms in the Hamiltonian can be written as

$$\alpha \sum_i P_{ix} - h \sum \sigma_i = - \sum (h - \alpha |P_{ix}|) \sigma_i \quad (3)$$

which shows that the value of $\alpha |P_{ix}|$ appears as an extra field. The mean value of $|P_{ix}|$ annulate the external field when $\alpha = 0.02$, and the convergency is the slowest for this value of $\alpha$.

To make the results clear, we made calculations as function of the temperature for different values of $\alpha$, when $\beta$ is zero - meaning that the magnetoelectric interaction is bigger than the electric one. After that, we calculated the minimum as function of $\beta$, when $\alpha$ is zero. The complete calculation, with both parameters different from zero gives a clear vision of the total behavior of our model.

Fig. 1 shows the results for the ferromagnetic case. The induced electric polarization appears at the same temperature as the magnetic transition, but shows important fluctuations at temperatures higher than $T_c$ as compared with those of the magnetic lattice.

### 2.3. Antiferromagnetic case

The antiferromagnetic case was treated similarly. It requires a negative value of $J$, but several changes in the other terms of the Hamiltonian are necessary. The magnetization of both sublattices will be coupled to the electric lattice opposed, in order to obtain the required ferroelectricity. Our results in this case are very similar to those above, and we lack here of space to show them completely. These results will be publish elsewhere, together with a more sophisticated model for the spin-lattice coupling.

### 3. Conclusions and perspectives.

The model presented above contains essentially the complete elements necessary to solve the magnetoelectric problem. Depending of the details in the spin-lattice (or magnetoelectric) Hamiltonian, the simulations will show different aspects of the physics involved. We are developing the approximation that corresponds with the creation of the electric moments, and the change of their values as functions of the local magnetization and strain. We believe that that approximation will simulate properly the case of the LiMPO$_4$ compounds.

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### 4. References

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