cecam Centre Européen de Calcul Atomique et Moléculaire	**Workshop Scientific Report** Please do not repeat the program (unless there were last-minute changes) or the initial description - we already have this material.
Title	Theory of Magnetoelectrics: Fundamentals and Applications
Organizers	Jorge Iniguez and Massimiliano Stengel

Scope of the workshop (one-two paragraphs)

This workshop focused on the physics of magnetoelectric materials, and on the current state of the art concerning fundamental theory, modeling and applications. The largest emphasis was given to first-principles methods; however, one of our main goals was to bring together researchers with different backgrounds, in order to foster a lively debate and encourage the exchange of ideas and perspectives. To that end, we also invited prominent experimental researchers to bring their point of view to the table and participate to the discussions.

More concretely, our aim was twofold. The first was to share the numerous novel theoretical methods that were developed during the past few years, and that are not yet "mainstream". This concerns in particular the orbital
magnetoelectric coupling, finite-field methods, linear-response approaches and effective Hamiltonians for magnetoelectric materials. The second was to critically discuss the latest developments in terms of materials design, and possibly get a flavor of what might be the most promising avenues for future research.

Main **outcomes** of key presentations (one page)

We believe it was the general impression among workshop participants that the quality of the presentations was extremelly high. Keeping in mind that most of the talks were truly outstanding, here we only list a selected few whose topics emerged as particularly novel or important.

We can group the key presentations in the workshop in three main categories:

- Fundamental theoretical developments

David Vanderbilt and Ivo Souza presented the latest developments in the fundamental theory of orbital magnetoelectric couplings, including effects related with the physics of topological insulators. This is becoming a very hot topic following recent experimental discoveries of exotic surface states in the prototype material Bi2Se3. The possibility of having a giant magnetoelectric response in such materials, and on-going computational attempts at verifying this prediction, was discussed.

Nicola Spaldin described pioneering attempts at defining and computing ferrotoroidal moments in magnetoelectrics. Beyond its fundamental interest, ferrotoroidicity could provide us with an additional order parameter to exploit in device applications. Prof. Spaldin's talk stimulated a lively and fruitful discussion on the challenges for a fundamental description of these effects. We are convinced our workshop will be the beginning of new activities and collaborations on this problem.

Juan Manuel Perez-Mato made a concise and illuminating presentation of group theory applied to magnetoelectric and multiferroic phenomena. He provided specific examples of how the use of group-theory tools (which have so far found limited application in this field) can facilitate the modeling and understanding of the underlying physics. After this talk, several opportunities for collaboration with other participants emerged.

- Applications to real materials / Predictions from theory:

Karin Rabe and Jorge Iniguez discussed specific strategies to obtain very large magnetoelectric responses by tuning (via epitaxial strain, chemical substitution, etc.) the properties of multiferroic perovskite oxides. These talks summarized very clearly the state of the art as regards lattice-mediated magnetoelectric couplings, and the prospects to obtain -- from this or other mechanisms -- materials that can be used in room-temperature devices.

Craig Fennie and Silvia Picozzi's contributions were especially important in that they described new forms of magnetostructural and magnetoelectric couplings in classes of materials that have so far received little attention
from our community. This included layered perovskite oxides, ortho-ferrites, sulfides, hybrid organic-inorganic materials, etc. These presentations were refreshing as they brought new ideas on the table; these possibilities are
likely to shape the activity in the field in the coming years.

- Experiment

We had very stimulating contributions from experimental colleagues. Among them, we would like to highlight those from Manuel Bibes (who discussed novel tunnel electro- and magneto-resistance effects that constitute a great
challenge for our first-principles community to model and explain) and J.F. Scott (who showed giant magnetoelectric effects at room temperature, obtained with a new class of multi-relaxor materials, which are likely to challenge theorists during the coming years).

Report on selected discussions (one page)
eg. Were there interesting hints for new research? for new developments? for collaborations?
1.- Modern theory of polarization

Following the talks by M. Stengel and A. Tagantsev, a lively discussion (mainly led by the two speakers, together with D. Vanderbilt and R. Resta) started about the proper use of electrical variables. A. Tagantsev challenged the possibility of defining the polarization P as a bulk property, and at the same time discussed some possible shortcomings of using the electric displacement D in Landau-theory models. This concerns directly the definition of magnetoelectric coefficients as second derivative of the free energy with respect to the magnetic and electric fields (or, alternatively, with respect to the magnetization and polarization). After the workshop, there was a follow-up discussion between M. Stengel, D. Vanderbilt and R. Resta about this topic, where these ideas were
further elaborated. This discussion was very fruitful -- at the time of writing, a preprint by R. Resta on magnetoelectric effects was posted on the cond-mat server (arXiv:1007.4186, 23 Jul 2010).

2.- Toroidal moments

As we mentioned above, a lively discussion followed the talk presented by N. Spaldin on ferrotoroidal moments in magnetoelectric materials. The main subject of the discussion was the fundamental definition of an origin-independent quantity. In some respect, the problem is analogous to that of polarization in crystalline insulators, where a rigorous definition necessarily implies some compromises (i.e. the polarization has to be understood as an intrinsically multivalued quantity, whose absolute value is not measurable). D. Vanderbilt pointed out that, unlike the case of electrical charge, the local spin moment is not quantized in a generic ferrotoroidic material, which makes the intrinsic multivaluedness much harder to control. P. Toledano pointed out that, for an order parameter to be "accepted" as a fundamental quantity together with magnetization and polarization, there must be a well-defined conjugate field associated to it, which seems to be missing in the ferrotoroidic case. This observation was further objected: structural degrees of freedom (e.g. rotation and tilts of the oxygen octahedra) are a valid counterexample of a well-defined order parameter without an obvious conjugate field. This discussion initiated follow-up discussions between the participants in order to clarify the connection between the current status of ab-initio theory and the definition of toroidal moment in phenomenological models.

3.- Effective modelling of temperature-driven effects in BiFeO3

Room temperature multiferroic BiFeO3 (BFO) was the focus of a number of talks. Specially relevant were those given by Kornev and Lisenkov, who described the development and application of first-principles effective
Hamiltonian methods to simulate the thermodynamic properties of BFO. The lively discussion in Igor Kornev's talk -- with Scott, Perez-Mato and Iniguez as main contributors from the public -- made it clear that BFO poses a
singular challenge to the effective Hamiltonian approach, which was previously successfully applied to "simple" ferroelectrics (like BaTiO3 or PbTiO3) by workshop participants Rabe and Vanderbilt. The discussion focused on the predictions for the temperature-driven phase transitions in BFO (the Heff approach reported by Kornev and Lisenkov predicts a I4/mcm phase above the ferroelectric transition, while recent experiments suggest a Pnma phase) and the stabilization of new phases in BFO films subject to epitaxial strain (the described Heff approach seems to require a novel magneto-structural coupling to comply with some experimental observations). These are issues of uttermost importance, as a detailed knowledge of the phase diagram of the material could
greatly facilitate progress towards applications. Specific suggestions as to how to overcome the current limitations of the effective models were debated.

4.- Future of the field

The workshop concluded with a discussion session, animated by K.M. Rabe, in which the future of the field was debated.

From the perspective of the fudamental theory, it became clear that there is still work to be done as regards a number of important issues. The most obvious examples are the above mentioned ones: the lack of a quantum-mechanical definition of ferrotoroidicity, the need for consensus as regards the electrical and magnetic variables used in ab initio and Landau theories, and the need for better effective models to simulate

Report on selected discussions (one page)
eg. Were there interesting hints for new research? for new developments? for collaborations?
1.- Modern theory of polarization

Following the talks by M. Stengel and A. Tagantsev, a lively discussion (mainly led by the two speakers, together with D. Vanderbilt and R. Resta) started about the proper use of electrical variables. A. Tagantsev challenged the possibility of defining the polarization P as a bulk property, and at the same time discussed some possible shortcomings of using the electric displacement D in Landau-theory models. This concerns directly the definition of magnetoelectric coefficients as second derivative of the free energy with respect to the magnetic and electric fields (or, alternatively, with respect to the magnetization and polarization). After the workshop, there was a follow-up discussion between M. Stengel, D. Vanderbilt and R. Resta about this topic, where these ideas were
further elaborated. This discussion was very fruitful -- at the time of writing, a preprint by R. Resta on magnetoelectric effects was posted on the cond-mat server (arXiv:1007.4186, 23 Jul 2010).

2.- Toroidal moments

As we mentioned above, a lively discussion followed the talk presented by N. Spaldin on ferrotoroidal moments in magnetoelectric materials. The main subject of the discussion was the fundamental definition of an origin-independent quantity. In some respect, the problem is analogous to that of polarization in crystalline insulators, where a rigorous definition necessarily implies some compromises (i.e. the polarization has to be understood as an intrinsically multivalued quantity, whose absolute value is not measurable). D. Vanderbilt pointed out that, unlike the case of electrical charge, the local spin moment is not quantized in a generic ferrotoroidic material, which makes the intrinsic multivaluedness much harder to control. P. Toledano pointed out that, for an order parameter to be "accepted" as a fundamental quantity together with magnetization and polarization, there must be a well-defined conjugate field associated to it, which seems to be missing in the ferrotoroidic case. This observation was further objected: structural degrees of freedom (e.g. rotation and tilts of the oxygen octahedra) are a valid counterexample of a well-defined order parameter without an obvious conjugate field. This discussion initiated follow-up discussions between the participants in order to clarify the connection between the current status of ab-initio theory and the definition of toroidal moment in phenomenological models.

3.- Effective modelling of temperature-driven effects in BiFeO3

Room temperature multiferroic BiFeO3 (BFO) was the focus of a number of talks. Specially relevant were those given by Kornev and Lisenkov, who described the development and application of first-principles effective
Hamiltonian methods to simulate the thermodynamic properties of BFO. The lively discussion in Igor Kornev's talk -- with Scott, Perez-Mato and Iniguez as main contributors from the public -- made it clear that BFO poses a
singular challenge to the effective Hamiltonian approach, which was previously successfully applied to "simple" ferroelectrics (like BaTiO3 or PbTiO3) by workshop participants Rabe and Vanderbilt. The discussion focused on the predictions for the temperature-driven phase transitions in BFO (the Heff approach reported by Kornev and Lisenkov predicts a I4/mcm phase above the ferroelectric transition, while recent experiments suggest a Pnma phase) and the stabilization of new phases in BFO films subject to epitaxial strain (the described Heff approach seems to require a novel magneto-structural coupling to comply with some experimental observations). These are issues of uttermost importance, as a detailed knowledge of the phase diagram of the material could
greatly facilitate progress towards applications. Specific suggestions as to how to overcome the current limitations of the effective models were debated.

4.- Future of the field

The workshop concluded with a discussion session, animated by K.M. Rabe, in which the future of the field was debated.

From the perspective of the fudamental theory, it became clear that there is still work to be done as regards a number of important issues. The most obvious examples are the above mentioned ones: the lack of a quantum-mechanical definition of ferrotoroidicity, the need for consensus as regards the electrical and magnetic variables used in ab initio and Landau theories, and the need for better effective models to simulate

To what extent were the **objectives** of the workshop achieved (strong points, weak points)? (one paragraph at least)

The basic objective of the workshop was to invite leaders from two disciplines to be exposed to each other's methods and ideas, and to interact and debate on the pros and cons of the methods. Of course it will take some time before actual ideas are implemented into new methods and codes. However, we feel that a very useful interaction took place, with healthy and lively debates.

Do you have suggestions for new workshops/tutorials/conferences on the topic?

We believe the basic theme of the workshop is far from exhausted, and worthy of another workshop in three of four years time, where some of the above methods have had more time to develop.