Coexistence of Ferroelectricity and Ferromagnetism

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The interplay between order parameters of different natures opens the door for designing new multifunctional devices whose properties can be manipulated with more than one physical field. For instance, the spin and orbital electronic degrees of freedom can order individually or simultaneously in a crystalline environment, producing abundant phases. In particular, orbital ordering can produce different symmetry-breaking states like orbital magnetism, ferroelectricity (FE), quadrupolar electric or magnetic ordering, and other multipolar orderings. The magnetoelectric multiferroics, such as $R(Fe,Mn)O_3$ and RMn_2O_5, are real examples of the multifunctional materials that combine distinct useful properties within a single system. Recent experimental studies on these multiferroic materials have revived interest in the magnetoelectric effect, i.e., the induction of polarization with an applied magnetic field and magnetization with an applied electric field. To date, materials that exhibit both ferromagnetism (FM) and FE are rare because the transition metal ion of typical perovskite ferroelectrics is in a nonmagnetic d_0 electronic configuration. Therefore, it is essential to explore alternative routes to the coexistence of the FM and FE.

Different mechanisms for FE involving electronic degrees of freedom have been proposed. There are those in which FE results from bond ordered states induced either by electron-phonon coupling (Peierls instability) or by pure electron-electron Coulomb interactions. In these cases, the ferroelectric state is clearly nonmagnetic due to the singlet nature of the covalent bonds. In contrast, considering a system of interacting *spinless* fermions with two atomic orbitals of *opposite* inversion symmetry (say the *d* and *f* orbitals), Portengen, et al. [1] predicted that permanent electric dipoles are induced by spontaneous *d-f* hybridization when particle-hole pairs (excitons) undergo a Bose-Einstein condensation. This result was confirmed in the strong coupling limit of an extended Falicov-Kimball spinless fermion model where both bands are dispersive [2]. It was also confirmed numerically in the intermediate coupling regime by using a constrained path Monte Carlo approach [3].

The critical question that naturally emerges is, "How do FE and magnetism interplay when real electrons, instead of spinless fermions, are considered?" We answered this question by proving that the mechanism proposed by Portengen,

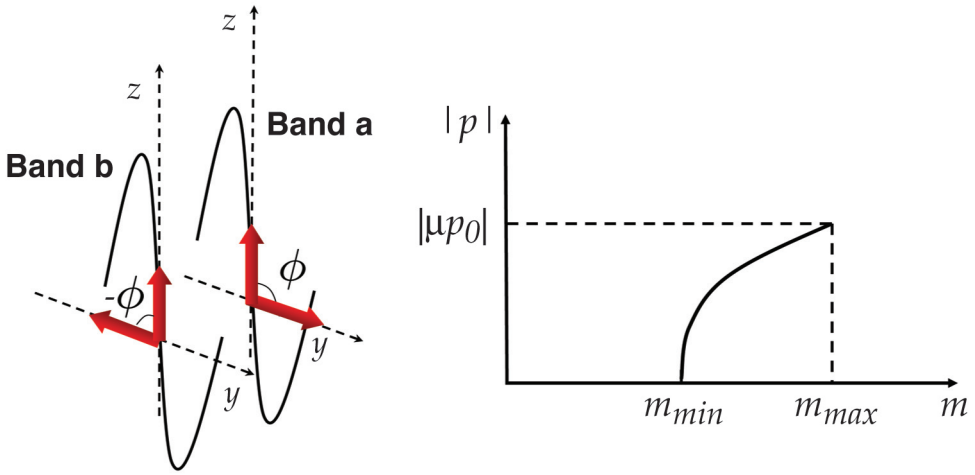

Fig. 1.
Evolution of the magnetization (arrows) on each band under the SO(4) transformation U_ϕ. The P vs M plot shows the change of the electric dipole moment P when the total magnetization M evolves from the minimum value, obtained for $\phi=\pi/2$, to the maximum value $M(=0)=N/2$, where N is the number of lattice sites.

et al. can coexist with magnetically ordered states. In this case, the single electron occupying the effective (say *d-f* hybridized) orbital simultaneously provides an electric and a magnetic-dipole moment, and the Coulomb repulsion is sufficient to generate a strong effective coupling between the electric and the magnetic dipoles.

We started from a two-band Hubbard Hamiltonian that included an interorbital on-site repulsive interaction U_{ab}. Like in the spinless fermion case, this interaction provided the "glue" for the formation of excitons. At quarter filling and in the strong coupling limit, we mapped the low energy spectrum of the two-band Hubbard model into an effective spin-pseudospin Hamiltonian. We proved that in the limit of large intraorbital repulsive interactions $U_{aa}, U_{bb} \to \infty$, the effective Hamiltonian has a ferromagnetic ground state that can be partially or fully saturated. By combining this result with the previous analysis for spinless fermions [2, 3, 4], we showed that FM and FE coexisted in that region of parameters, and a divergent magnetoelectric was demonstrated by using the SO(4) symmetry of the original Hamiltonian (see Fig. 1). Our conclusions were reinforced by a semiclassical and a numerical computation of the zero temperature ($T = 0$) phase diagram of original Hamiltonian that went beyond the limiting case $U_{aa}, U_{bb} \to \infty$. These computations allowed us to sketch the $T = 0$ phase diagram for the effective Hamiltonian (Fig. 2). We note the abundance of phases.

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[1] T. Portengen, et al., Phys. Rev. Lett. 76, 3384 (1996); *Phys. Rev.* **54**, 17452 (1996).
[2] C.D. Batista, *Phys. Rev. Lett.* **89**, 166403 (2002).
[3] C.D. Batista, et al., *Phys. Rev. Lett.* **92**, 187601 (2004).
[4] W.G. Yin et al., *Phys. Rev. B* **8**, 075111 (2003).

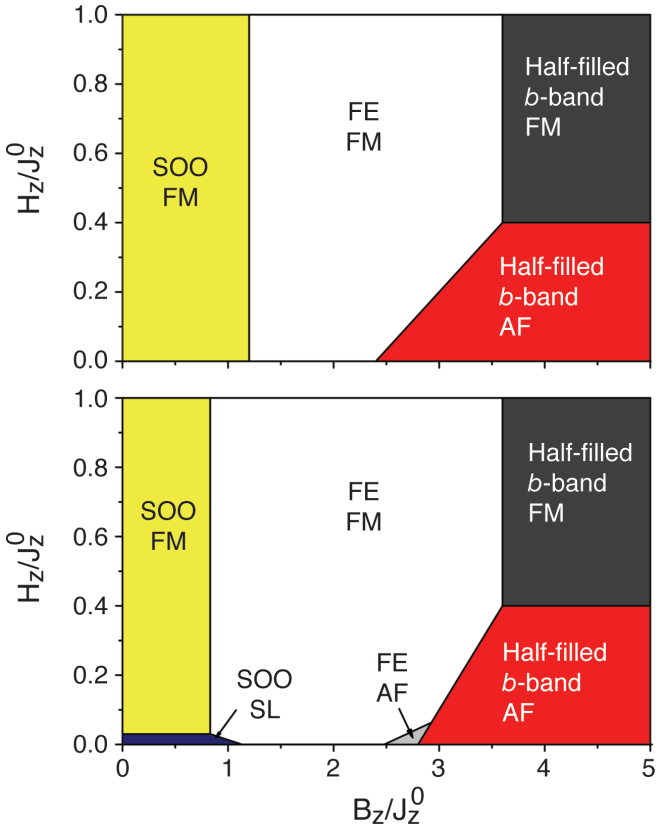

Fig. 2. ***Zero temperature phase diagram of the two-dimensional version of the effective Hamiltonian plus a Zeeman term computed in the spin-wave approximation (top) and by exact diagonalization of a 4 × 4 cluster (bottom).***