Electronic mechanism for the coexistence of ferroelectricity and ferromagnetism

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We study the strong-coupling limit of a two-band Hubbard Hamiltonian that includes an interorbital on-site repulsive interaction U_{ab}. When the two bands have opposite parity and are quarter filled, we prove that the ground state is simultaneously ferromagnetic and ferroelectric for intraorbital Coulomb interactions $U_{aa}, U_{bb} \gg U_{ab}$. We also show that this coexistence leads to an enormous magnetoelectric effect.

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INTRODUCTION

The interplay between order parameters of different nature 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 to produce different phases. In particular, orbital ordering can produce symmetry-breaking states like orbital magnetism, ferroelectricity (FE), quadrupolar electric or magnetic ordering, and other multipolar orderings. The magnetoelectric multiferroics, such as $R(\mathrm{Fe},\mathrm{Mn})\mathrm{O}_3$ and $R\mathrm{Mn}_2\mathrm{O}_5$,[1–7] are real examples of materials that combine distinct useful properties within a single system. Recent studies of 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 FM and FE.[8]

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[9] (Peierls instability) or by pure electron-electron Coulomb interactions.[10] In these cases, the ferroelectric state is clearly nonmagnetic due to the singlet nature of the covalent bonds. Recently, it was suggested that FE and magnetic order might coexist in near-half-doped manganites in the form of a combined site- and bond-centered charge order of the Mn d electrons.[11] 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.*[12] 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.[13] It was also confirmed numerically in the intermediate-coupling regime by using a constrained path Monte Carlo approach.[14]

The purpose of this paper is to show that the mechanism proposed by Portengen *et al.*[12–15] can coexist with magnetically ordered states when real electrons, instead of spinless fermions, are considered. 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 combined with the Pauli exclusion principle generate a strong coupling between them. More specifically, we consider a two-band Hubbard Hamiltonian that includes on-site intraorbital repulsive interactions U_{aa}, U_{bb} and an interorbital interaction U_{ab}.[16] As in the spinless fermion case, U_{ab} provides the "glue" for the formation of excitons. In the strong-coupling limit and at quarter filling (one electron per site), we prove that the ground state is simultaneously FE and FM for $U_{aa}, U_{bb} \gg U_{ab}$. Furthermore, we demonstrate from symmetry considerations that this coexistence leads to a divergent magnetoelectric effect. Our conclusions are reinforced by a semiclassical and a numerical computation of the zero-temperature phase diagram beyond the limiting case of $U_{aa}, U_{bb} \gg U_{ab}$.

HAMILTONIAN

Our two-band Hubbard model on a D-dimensional hypercubic lattice is[16]

$$H = \sum_{\mathbf{i},\eta,\nu,\nu',\sigma} t_{\nu\nu'}(f^{\dagger}_{\mathbf{i}\nu\sigma} f_{\mathbf{i}+\hat{\mathbf{e}}_\eta \nu'\sigma} + f^{\dagger}_{\mathbf{i}+\hat{\mathbf{e}}_\eta \nu'\sigma} f_{\mathbf{i}\nu\sigma}) + \sum_{\mathbf{i}} U_{ab} n^a_{\mathbf{i}} n^b_{\mathbf{i}} + \sum_{\mathbf{i},\nu} U_{\nu\nu} n^{\nu}_{\mathbf{i}\uparrow} n^{\nu}_{\mathbf{i}\downarrow} + \sum_{\mathbf{i},\nu} \epsilon_\nu n^{\nu}_{\mathbf{i}}, \quad (1)$$

where $\eta=\{x,y,z,\ldots\}$, $\nu=\{a,b\}$, $n^{\nu}_{\mathbf{i}\sigma}=f^{\dagger}_{\mathbf{i}\nu\sigma} f_{\mathbf{i}\nu\sigma}$, $n^{\nu}_{\mathbf{i}}=\Sigma_\sigma n^{\nu}_{\mathbf{i}\sigma}$, and $n_{\mathbf{i}}=\Sigma_\nu n^{\nu}_{\mathbf{i}}$. Since the two orbitals a and b have opposite parity under spatial inversion, the interband hybridization term must be odd under this operation: $t_{ab}=-t_{ba}$. In addition, the intraband hoppings t_{aa} and t_{bb} will have *opposite* signs in general. The local spin and pseudospin operators are given by the expressions

$$s^{\mu}_{\mathbf{i}\nu} = \frac{1}{2}\sum_{\alpha\alpha'} f^{\dagger}_{\mathbf{i}\nu\alpha} \sigma^{\mu}_{\alpha\alpha'} f_{\mathbf{i}\nu\alpha'}, \quad \tau^{\mu}_{\mathbf{i}\sigma} = \frac{1}{2}\sum_{\nu\nu'} f^{\dagger}_{\mathbf{i}\nu\sigma} \sigma^{\mu}_{\nu\nu'} f_{\mathbf{i}\nu'\sigma}, \quad (2)$$

where σ^{μ} are the Pauli matrices with $\mu=\{x,y,z\}$. The total spin and pseudospin per site are $s^{\mu}_{\mathbf{i}}=\Sigma_\nu s^{\mu}_{\mathbf{i}\nu}$ and $\tau^{\mu}_{\mathbf{i}}=\Sigma_\sigma \tau^{\mu}_{\mathbf{i}\sigma}$. The pseudospin component $\tau^{x}_{\mathbf{i}}$ is proportional to the on-site hybridization. Since the a and b orbitals have opposite parity, the local electric dipole moment is $\mathbf{p_i}=\boldsymbol{\mu}\tau^{x}_{\mathbf{i}}$, where $\boldsymbol{\mu}$ is the dipole matrix element between the a and b orbitals.[13,14]

Symmetry is a useful concept for describing the coexistence of different order parameters.[17] For $t_{aa}=\pm t_{bb}$, $t_{ab}=t_{ba}=0$, $\epsilon_a-U_{aa}/2=\epsilon_b-U_{bb}/2$, and $U_{aa}=U_{ab}=U_{bb}$, H is invariant under a $U(1)\otimes SU(4)$ symmetry group. The U(1) symmetry corresponds to the conservation of the total number of particles. The generators of the SU(4) symmetry group are the three components of the total spin $s_T^\mu=\Sigma_{\mathbf{i}}s_{\mathbf{i}}^\mu$ and pseudospin $\tau_T^\mu=\Sigma_{\mathbf{i}}\tau_{\mathbf{i}}^\mu$ plus the nine operators

$$\pi^{\mu\mu'}=\frac{1}{2}\sum_{\mathbf{i},\alpha\nu\alpha'\nu'} f_{\mathbf{i}\nu\alpha}^\dagger \sigma_{\nu\nu'}^\mu \sigma_{\alpha\alpha'}^\mu f_{\mathbf{i}\nu'\alpha'}. \tag{3}$$

The total spin is conserved for any set of parameters. If we just impose the condition $t_{ab}=t_{ba}=0$, the symmetry group of H is reduced to the the subgroup $U(1)\times U(1)\times SO(4)$. The six generators of the SO(4) group are the three components of total spin s_T^μ and the three operators $\pi^{z\mu}$. This symmetry arises from separate total spin and charge conservation of each band, as the two bands are coupled only by the Coulomb interaction U_{ab}. The symmetry operators $s_T^\mu\pm\pi^{z\mu}$ are the generators of global spin rotations on each individual band (with the + sign for the a band and the − sign for the b band).

STRONG-COUPLING LIMIT

We will consider from now on the quarter-filled case $n_a+n_b=1$, where n_a and n_b are the particle densities of the bands a and b. When $U_{aa},U_{bb},U_{ab}\gg|t_{\nu\nu'}|$, the low-energy spectrum of H can be mapped to an effective spin-pseudospin Hamiltonian by means of a canonical transformation that eliminates the linear terms in $t_{\nu\nu'}$:

$$\begin{aligned}H_{\text{eff}}=&\sum_{\mathbf{i},\eta}\left(\sum_\mu J_\mu \tau_{\mathbf{i}}^\mu \tau_{\mathbf{i}+\mathbf{e}_\eta}^\mu + J_{xz}(\tau_{\mathbf{i}}^z\tau_{\mathbf{i}+\mathbf{e}_\eta}^x-\tau_{\mathbf{i}}^x\tau_{\mathbf{i}+\mathbf{e}_\eta}^z)\right)H_{\mathbf{i},\eta}^H\\&+\sum_{\mathbf{i},\eta}\left(\frac{J_0}{2}+J_1(\tau_{\mathbf{i}}^z+\tau_{\mathbf{i}+\mathbf{e}_\eta}^z)-J_2(\tau_{\mathbf{i}}^x-\tau_{\mathbf{i}+\mathbf{e}_\eta}^x)\right)\left(H_{\mathbf{i},\eta}^H-\frac{1}{2}\right)\\&+\sum_{\mathbf{i},\eta}[J_z'\tau_{\mathbf{i}}^z\tau_{\mathbf{i}+\mathbf{e}_\eta}^z+J_{xz}'(\tau_{\mathbf{i}}^z\tau_{\mathbf{i}+\mathbf{e}_\eta}^x-\tau_{\mathbf{i}}^x\tau_{\mathbf{i}+\mathbf{e}_\eta}^z)]+\sum_{\mathbf{i}}B_z\tau_{\mathbf{i}}^z,\end{aligned}$$

where $H_{\mathbf{i},\eta}^H=\mathbf{s}_{\mathbf{i}}\cdot\mathbf{s}_{\mathbf{i}+\mathbf{e}_\eta}+\frac{1}{4}$ and

$$J_z=\frac{4(t_{bb}^2-t_{ab}^2)}{U_{bb}}+\frac{4(t_{aa}^2-t_{ab}^2)}{U_{aa}},$$

$$J_x=\frac{8(t_{bb}t_{aa}-t_{ab}^2)}{U_{ab}},\quad J_y=\frac{8(t_{bb}t_{aa}+t_{ab}^2)}{U_{ab}},$$

$$J_{xz}=4t_{ab}\left(\frac{(t_{aa}+t_{bb})}{U_{ab}}-\frac{t_{aa}}{U_{aa}}-\frac{t_{bb}}{U_{bb}}\right),$$

$$J_0=\frac{2(t_{bb}^2+t_{ab}^2)}{U_{bb}}+\frac{2(t_{aa}^2+t_{ab}^2)}{U_{aa}},\quad J_1=\frac{2t_{bb}^2}{U_{bb}}-\frac{2t_{aa}^2}{U_{aa}},$$

$$J_2=2t_{ab}\left(\frac{(t_{aa}-t_{bb})}{U_{ab}}+\frac{t_{aa}}{U_{aa}}-\frac{t_{bb}}{U_{bb}}\right),$$

$$J_z'=\frac{2(t_{aa}^2+t_{bb}^2-2t_{ab}^2)}{U_{ab}}-\frac{J_z}{2},$$

$$J_{xz}'=2t_{ab}\left(\frac{(t_{aa}+t_{bb})}{U_{ab}}+\frac{t_{aa}}{U_{aa}}+\frac{t_{bb}}{U_{bb}}\right), \tag{4}$$

and $B_z=\epsilon_a-\epsilon_b+(U_{bb}-U_{aa})/2$. In this limit, because double occupancy is forbidden in the low-energy Hilbert space of H_{eff}, both $\mathbf{s}_{\mathbf{i}}$ and $\tau_{\mathbf{i}}$ belong to the $S=1/2$ representation of the su(2) algebra. The first two terms of H_{eff} couple the spin and orbital degrees of freedom. As usual, the Heisenberg antiferromagnetic interaction $H_{\mathbf{i},\eta}^H$ is a direct consequence of the Pauli exclusion principle. On the other hand, the anisotropic Heisenberg-like pseudospin-pseudospin interaction reflects the competition between an excitonic crystallization or staggered orbital ordering (SOO) induced by the Ising term, and a Bose-Einstein condensation of excitons induced by the XY term.[13,15] The first term of H_{eff} shows explicitly that the amplitude of the excitonic kinetic energy (or XY pseudospin) term gets maximized when the excitons are in a fully polarized ferromagnetic spin state. However, antiferromagnetism (AFM) is clearly favored by the second term.

LARGE-U_{aa}, U_{bb} LIMIT

We will first prove that there are partially and fully polarized ferromagnetic ground states of H_{eff} in the limit of $U_{aa},U_{bb}\to\infty$ and $t_{ab}=0$, and that the total spin and magnetization s_T take the values $\tau_T^z\le s_T\le N/2$. After proving this result, we will show that these ferromagnetic solutions are also ferroelectric for $B_z^{c1}\le B_z<B_z^{c2}$ and, using the SO(4) symmetry, we will derive an exact expression for ground-state electric polarization as a function of the magnetization.

Since J_0 and J_1 vanish in this limit, H_{eff} is reduced to

$$\bar{H}_{\text{eff}}=\sum_{\langle\mathbf{i},\mathbf{j}\rangle}[J_z'\tau_i^z\tau_j^z+J_\perp(\tau_{\mathbf{i}}^x\tau_{\mathbf{j}}^x+\tau_{\mathbf{i}}^y\tau_{\mathbf{j}}^y)H_{\mathbf{i},\mathbf{j}}^H]+B_z\sum_{\mathbf{i}}\tau_{\mathbf{i}}^z,$$

where the angular brackets indicate that the sum is over nearest neighbors, $J_\perp=J_x=J_y$, and $H_{\mathbf{i},\mathbf{j}}^H=\mathbf{s}_{\mathbf{i}}\cdot\mathbf{s}_{\mathbf{j}}+\frac{1}{4}$. To prove our statement we will use a basis of eigenstates of the local operators $\tau_{\mathbf{i}}^z$ and $s_{\mathbf{i}}^z$: $\{|\tau_1^z\cdots\tau_N^z\rangle\otimes|s_1^z\cdots s_N^z\rangle\}$, where N is the total number of sites. The off-diagonal matrix elements of $\bar{H}_{\text{eff}}$,

$$\begin{aligned}&\langle s_N'^z\cdots s_1'^z|\otimes\langle\tau_N'^z\cdots\tau_1'^z|\bar{H}_{\text{eff}}|\tau_1^z\cdots\tau_N^z\rangle\otimes|s_1^z\cdots s_N^z\rangle\\&\quad=2J_\perp\sum_{\langle\mathbf{i},\mathbf{j}\rangle}\langle\tau_N'^z\cdots\tau_1'^z|H_{\mathbf{i},\mathbf{j}}^{xy}|\tau_1^z\cdots\tau_N^z\rangle\langle s_N'^z\cdots s_1'^z|H_{\mathbf{i},\mathbf{j}}^H|s_1^z\cdots s_N^z\rangle,\end{aligned} \tag{5}$$

are nonpositive because $J_\perp<0$, and the matrix elements of $H_{\mathbf{i},\mathbf{j}}^{xy}=\tau_{\mathbf{i}}^x\tau_{\mathbf{j}}^x+\tau_{\mathbf{i}}^y\tau_{\mathbf{j}}^y$ and $H_{\mathbf{i},\mathbf{j}}^H$ are explicitly non-negative. According to the generalized Perron theorem (see, for instance, Ref. 18), there is one ground state of $\bar{H}_{\text{eff}}$,

$$|\Psi^g\rangle=\sum_{\{\tau^z\}}|\tau_1^z\cdots\tau_N^z\rangle\otimes\sum_{\{s^z,s_T^z=0\}}a_{\{\tau^z\},\{s^z\}}|s_1^z\cdots s_N^z\rangle, \tag{6}$$

such that all the amplitudes $a_{\{\tau^z\},\{s^z\}}$ are non-negative ($\{\tau^z\}$ and $\{s^z\}$ denote all the possible configurations of $\tau_{\mathbf{i}}^z$ and $s_{\mathbf{i}}^z$).

We can rewrite $|\Psi^g_{s^z_T=0}\rangle$ in the following way:

$$|\Psi^g\rangle = \sum_{\{\tau^z\}'} b_{\{\tau^z\}} |\tau^z_1 \cdots \tau^z_N\rangle \otimes |\Phi\{\tau^z\}\rangle, \tag{7}$$

with

$$|\Phi\{\tau^z\}\rangle = \frac{1}{b_{\{\tau^z\}}} \sum_{\{s^z, s^z_T=0\}} a_{\{\tau^z\},\{s^z\}} |s^z_1 \cdots s^z_N\rangle,$$

$$b_{\{\tau^z\}} = \sqrt{\sum_{\{s^z, s^z_T=0\}} a^2_{\{\tau^z\},\{s^z\}}}. \tag{8}$$

The set $\{\tau^z\}'$ corresponds to all the configurations of the $\tau^z_{\mathbf{i}}$ variables such that $b_{\{\tau^z\}}>0$. Note that each spin state $|\Phi\{\tau^z\}\rangle$ is normalized and $\Sigma_{\mathbf{i}} b^2_{\{\tau^z\}}=1$ because $|\Psi^g\rangle$ is also normalized. The ground-state energy of $\bar{H}_{\text{eff}}$ is

$$\begin{aligned}\langle\Psi^g|\bar{H}_{\text{eff}}|\Psi^g\rangle = \langle\Psi^g|\sum_{\langle\mathbf{i},\mathbf{j}\rangle} J_z \tau^z_i \tau^z_j + B_z \sum_{\mathbf{i}} \tau^z_{\mathbf{i}}|\Psi^g\rangle \\ + 2J_\perp \sum_{\langle\mathbf{i},\mathbf{j}\rangle} \sum_{\{\tau^z,\tau'^z\}'} b_{\{\tau^z\}} b_{\{\tau'^z\}} \\ \times \langle\tau'^z_N \cdots \tau'^z_1 | H^{xy}_{\mathbf{i},\mathbf{j}} | \tau^z_1 \cdots \tau^z_N\rangle \times \langle\Phi\{\tau'^z\}| \\ \times |H^H_{\mathbf{i},\mathbf{j}}|\Phi\{\tau^z\}\rangle.\end{aligned} \tag{9}$$

$H^H_{\mathbf{i},\mathbf{j}}$ is a bounded operator with eigenvalues $\pm 1/2$. Therefore $\langle\Phi\{\tau'^z\}|H^H_{\mathbf{i},\mathbf{j}}|\Phi\{\tau^z\}\rangle \leq 1/2$ and the equality holds for any pair of nearest-neighbor sites $(\mathbf{i},\mathbf{j})$ if $|\Phi\{\tau^z\}\rangle = |s^z_T, s_T = N/2\rangle$ where $|s^z_T, s_T = N/2\rangle$ is the state with maximum total spin (fully polarized) and $\Sigma_{\mathbf{i}} s^z_{\mathbf{i}} = s^z_T$. Hence, expression (9) is minimized for the fully polarized spin configuration which means that there is a family of ground states $|\Psi^g\rangle$ that have maximum total spin s_T and different values of s^z_T:

$$|\Psi^g\rangle = \sum_{\{\tau^z\}'} b_{\{\tau^z\}} |\tau^z_1 \cdots \tau^z_N\rangle \otimes |s^z_T, s_T = N/2\rangle. \tag{10}$$

This proves that there is a fully polarized ferromagnetic ground state of $\bar{H}_{\text{eff}}$. Note that the proof is valid in any dimension and is also valid for nonbipartite lattices. When we restrict $\bar{H}_{\text{eff}}$ to the subspace with maximum total spin ($s_T = N/2$), the operator $\left(\mathbf{S_i}\cdot\mathbf{S_j} + \frac{1}{4}\right)$ is replaced by $1/2$ and the restricted Hamiltonian $\bar{H}^{\text{FM}}_{\text{eff}}$ becomes exactly the same as the one obtained in Ref. 13 for the strong-coupling limit of a spinless extended Falicov-Kimball model.[19] As shown in Refs. 13 and 15, the quantum phase diagram of $\bar{H}^{\text{FM}}_{\text{eff}}$ contains a ferroelectric phase for $B^{c1}_z \leq B_z < B^{c2}_z$, i.e., for a nonzero value of τ^z_T.

The SO(4) symmetry of $\bar{H}_{\text{eff}}$ implies that the ground-state degeneracy is higher than the $2s_T+1 = N+1$ multiplet obtained from the global SU(2) spin rotations. Ground states with different total spin can also be obtained by making different global spin rotations for the bands a and b. The spins of each individual band will remain fully polarized under these transformations, but the relative orientation between spins of different bands will change. In particular, the minimum total spin will occur when spins in different bands are "anti-aligned," i.e., $s_T = \tau^z_T$. This implies that the total spin of the ground state can take the values $\tau^z_T \leq s_T \leq N/2$.

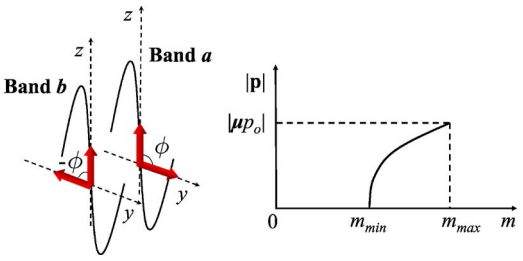

FIG. 1. (Color online) Evolution of the magnetization (arrows) on each band under the SO(4) transformation $U(\phi)$. The $|\mathbf{p}|$ vs $\mathbf{m}$ plot shows the change of the electric dipole moment when the total magnetization evolves from the minimum value, obtained for $\phi = \pi/2$, to the maximum value $m(\phi=0) = 1/2$.

To derive the magnetoelectric effect, we define $|\chi\rangle$ as the fully polarized ferromagnetic and ferroelectric state obtained for $B^{c1}_z \leq B_z < B^{c2}_z$. Then, the average magnetization and electric dipole moment per site, m and $\mathbf{p}$, are given by

$$m = \frac{\sqrt{\langle\chi|\mathbf{s}^2_T|\chi\rangle}}{N} = \frac{1}{2}\sqrt{1+\frac{2}{N}}, \quad \mathbf{p} = \boldsymbol{\mu} p_0, \tag{11}$$

where $p_0 = \langle\chi|\tau^x_T|\chi\rangle/N$. A given ground state in the SO(4) multiplet can be expressed as $|g_u\rangle = U|\chi\rangle$, where U is an element of the SO(4) group. In particular, choosing the set of transformations $U(\phi) = \exp[i\phi\Sigma_{\mathbf{j}}(S^x_{\mathbf{j}a} - S^x_{\mathbf{j}b})]$ we get for $\mathbf{p}(\phi) = \boldsymbol{\mu}\langle g_{u(\phi)}|\tau^x_T|g_{u(\phi)}\rangle/N$ and $m(\phi) = \sqrt{\langle g_{u(\phi)}|\mathbf{s}^2_T|g_{u(\phi)}\rangle}/N$:

$$m(\phi) = \frac{1}{2}\sqrt{1 - 4n_a n_b \sin^2\phi + \frac{2}{N}},$$

$$\mathbf{p}(\phi) = \boldsymbol{\mu} p_0 \cos\phi, \tag{12}$$

where we have used that $\mathbf{s}_T = \mathbf{s}_{aT} + \mathbf{s}_{bT}$ and that $U(\phi)$ rotates the vectors $\mathbf{s}_{aT}$ and $\mathbf{s}_{bT}$ around the x axis by angles ϕ and $-\phi$, respectively. For the second relation, we have used that

$$U^\dagger(\phi)\tau^x_T U(\phi) = \cos\phi\,\tau^x_T - \sin\phi\,\pi^{yx}, \tag{13}$$

and $\langle\chi|\pi^{yx}|\chi\rangle = 0$. Note that the transformation $U^\dagger(\phi)$ is generated by $\pi^{z\mu}$.[20] For $\phi = \pi/2$, the magnetizations of the a and b bands have opposite signs (see Fig. 1) and the total magnetization per site is minimized: $m_{\min} = m(\pi/2) = |n_a - n_b|/2$ (we have taken the thermodynamic limit $N \to \infty$). The electric polarization can be expressed as a function of m by combining Eqs. (12):

$$|\mathbf{p}| = 2|\boldsymbol{\mu} p_0| \frac{\sqrt{m^2 - m^2_{\min}}}{\sqrt{1 - 4m^2_{\min}}}. \tag{14}$$

The electric dipole moment is zero for $m = m_{\min}$ and it increases as $\sqrt{m - m_{\min}}$, implying that the derivative $d|\mathbf{p}|/dm$ diverges at $m = m_{\min}$ as $1/\sqrt{m - m_{\min}}$. This important result shows that the interplay between the spin and the orbital degrees of freedom can produce a singular magnetoelectric effect (see Fig. 1).

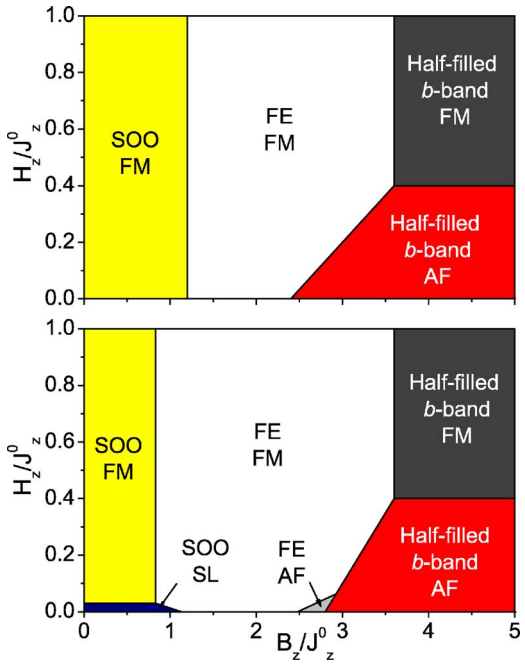

FIG. 2. (Color online) Zero-temperature phase diagram of the two-dimensional version of H_{eff} plus a Zeeman term $H_z s^z_T$ computed in the spin-wave approximation (top) and by Lanczos diagonalization of a 4×4 cluster (bottom). The parameter values are $J_0=0.2J^0_z$, $J_x=J_y=-1.6J^0_z$, $J_1=-0.1J^0_z$, and $J^0_z=J'_z+J_z/2$.

It is important to verify the stability of the coexisting ferroelectric and ferromagnetic phases away from the limit considered above. For this purpose, we computed the $T=0$ diagram of the two dimensional version of H_{eff} plus a Zeeman term $H_z s^z_T$ in a spin-wave approximation (top of Fig. 2) and by exact diagonalization of a 4×4 cluster (bottom of Fig. 2). In this case, the values of U_{aa} and U_{bb} are finite ($J_0=0.2J^0_z$), and the coexistence of FM and FE obtained for $H_z=0$ and $B^{c1}_z \le B_z < B^{c2}_z$ is still present (see Fig. 2). For $B_z < B^{c1}_z$ and $H_z=0$, the spin-wave phase diagram exhibits coexistence of FM and SOO.[13,15] The FM ordering is replaced by a spin liquid (short-ranged spin-spin correlations) in the phase diagram computed by exact diagonalization, indicating that quantum fluctuations play a major role in that regime of parameters. As expected, for large enough B_z the system becomes an antiferromagnetic Mott insulator (one band is a half filled and the other one is empty). The exact diagonalization shows again that quantum fluctuations generate an intermediate phase between the FE-FM state and the Mott insulator in which FE and AFM coexist (see bottom of Fig. 2). For high values of H_z, the system is fully polarized and both $T=0$ phase diagrams coincide with the one obtained in Refs. 13 and 15.

Figure 3 shows a comparison between the analytical expression for the electric polarization as a function of the magnetization given by Eq. (14) (valid in the thermodynamic limit) and the corresponding numerical results obtained from the Lanczos diagonalization of a 4×4 cluster. The good agreement between both results indicates that size effects are small, at least for the most relevant FM and FE phase.

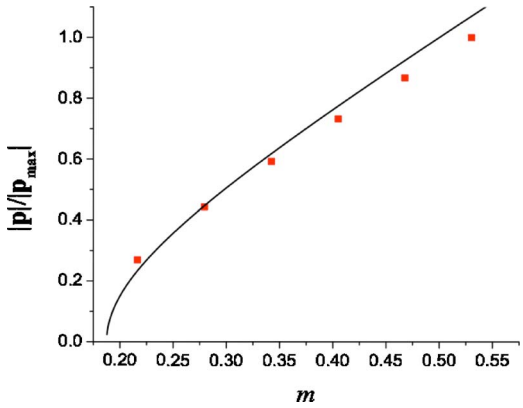

FIG. 3. (Color online) Comparison between the $|\mathbf{p}|$ vs $\mathbf{m}$ curve predicted by Eq. (14) (full line) and the result obtained by the Lanczos diagonalization of a 4×4 cluster (squares) for the FM and FE phase (see Fig. 2).

Ferromagnetism can be further stabilized by the inclusion of the *ferromagnetic* on-site interorbital exchange interaction. For example, the intra-atomic $4f$-$5d$ exchange interaction is about 0.2 eV in EuB_6.[22,23] The other important aspect to consider is the role of a finite interband hybridization t_{ab}. Exact diagonalization results[21] show that the lowest total spin ground state, $s_T=\tau^z_T$ or $m=m_{\text{min}}$, is the one stabilized after the inclusion of a small t_{ab} term. According to Eq. (14), this unsaturated ferromagnetic state gives rise to a divergent magnetoelectric effect. In this situation, a small increase in the magnetization produced by an applied magnetic field will generate a large increase of the electric dipole moment in the way depicted in Fig. 1.

In summary, we have shown that the electron-electron Coulomb interaction can produce coexisting FM and FE. Both phases arise simultaneously from the condensation of excitons or particle-hole pairs that exist in two bands with opposite parity under spatial inversion. The coexistence requires the presence of large intraorbital Coulomb interactions to reduce the strength of the antiferromagnetic interaction. We have also shown that the coexistence of FE and FM leads to a divergent magnetoelectric effect. In the proximity of the ferroelectric-ferromagnetic instability, a small magnetic field can produce an enormous change in the electric polarization.

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