Ferromagnetism in Itinerant Two-Dimensional t_{2g} Systems

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Motivated by the recent indications of ferromagnetism in transition metal oxide heterostructures, we propose a possible mechanism to generate ferromagnetism for itinerant t_{2g} systems in two spatial dimensions that does not rely on the coupling between local moments and conduction electrons. We particularly emphasize the orbital nature of different bands and show that when the Fermi level lies near the bottom of the upper bands, a nonperturbative interaction effect due to the quasi-one-dimensional nature of the upper bands may drive a transition to a state in which the upper bands are ferromagnetically polarized. In the quasi-one-dimensional limit, the full thermodynamics may be obtained exactly. We discuss the connection between our mechanism with several itinerant t_{2g} systems that may have ferromagnetic instabilities.

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Possible ferromagnetism at polar interfaces between $SrTiO_3$ (STO) and other oxides such as $LaAlO_3$ (LAO) or $GdTiO_3$ (GTO) [1–6] has raised considerable excitement. Such ferromagnetism is remarkable as the electrons are believed to reside nearly completely in the t_{2g} bands of the STO, where there are no localized partially filled shells to form local moments. Ferromagnetism in purely itinerant systems, while envisioned long ago [7], is quite rare in practice; most examples may be at least partially attributed to local moments formed by partially filled d-shells (and often other delocalized electrons).

In this Letter, we discuss the possibility of ferromagnetism in t_{2g} systems of this type. In a typical metallic state, ferromagnetism is unfavorable because of the kinetic energy cost. This is believed to be overcome at very low density, where the dimensionless interelectron distance $r_s \gtrsim 30$ [8,9]), and also very close to the Mott metal-insulator transition, when the electron filling is close to an integer. In the former regime, ferromagnetism and indeed metallicity as well are extremely fragile to disorder and may be disregarded in almost all practical situations. The latter situation might be thought to apply to the aforementioned LAO/STO and GTO/STO interfaces, in which there is an intrinsic mechanism for high carrier density: the polar discontinuity [10]. LAO and GTO have a structure of polar (001) layers: $La^{3+}O^{2-}$ has a net charge of $+1$ per unit cell, while $Al^{3+}(O^{2-})_2$ has a net charge of -1 per unit cell (the same counting holds for GTO). STO by contrast is nonpolar. At an ideal (i.e., without atomic reconstruction or compensating defects) interface between two such materials, an electron gas is predicted to arise with a carrier density of half an electron per two-dimensional unit cell. This translates, using the unit cell of STO, into a two-dimensional carrier density of $n = 3.5 \times 10^{14}$ cm^{-2}, which is extremely large by semiconductor standards. However, this still corresponds to a fractional Ti site occupation $x < 0.5$, and probably more properly $x < 0.2$, taking into account the spread of the electrons normal to the interface [11–14] Modern computational studies have put strong restrictions on ferromagnetism due to Mott physics in Hubbard models [15], the most recent studies arguing it is absent in the two-dimensional Hubbard model for fractional site occupation $x \lesssim 0.7$, even when the on-site Hubbard interaction $U \to \infty$ [16,17]. STO two-dimensional electron gases (2DEGs) are well below this degree of site occupation, so Mott physics cannot be invoked to explain ferromagnetism.

Instead, we propose here that an unusual enhanced tendency to ferromagnetism may occur due to the quasi-one-dimensionality of certain bands in these materials, which in turn arises due to the directionality of the t_{2g} orbitals involved. The enhanced tendency to magnetism is a nonperturbative effect of the Hubbard U interaction, which leads to strong scattering in one-dimensional subbands with low filling. The nonperturbative effects can be controlled by virtue of exact results and bosonization methods which are particular to one-dimensional problems. Due to the nonperturbative effect, ferromagnetism is induced by even very weak atomic Hund's exchange on the Ti atom (see below). We argue that the ferromagnetism survives in sufficiently anisotropic two-dimensional systems. It occurs only for low filling of the xz/yz subbands, where the majority of the polarization resides. The central result of our calculations is summarized by the phase diagram in Fig. 1.

For our discussion, we will require a few particulars of the conduction band states in STO, which are well established. The low-lying octahedral t_{2g} crystal field levels of Ti comprise yz, xz, and xy orbitals. Owing to its directionality, hopping t in the plane of a given orbital is much larger than the hopping t' normal to the plane (values in the literature are in the range $0.03 < t'/t < 0.15$ [11]). Thus

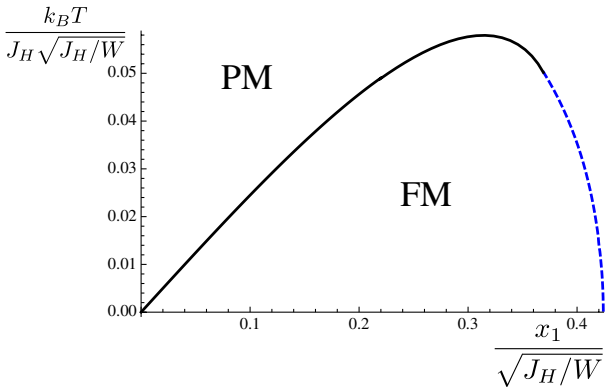

FIG. 1 (color online). Phase boundary in the one-dimensional limit, $t' = 0$. Here x_1 is the occupation per site of the xz or yz orbitals, J_H is the renormalized Hund's coupling (see text), W is an energy scale of order the hopping, and it is assumed that x_1, $J_H/W \ll 1$. Solid and dashed lines denote continuous and first-order transitions, respectively.

in a bulk system with cubic symmetry, there are three bands each of which disperses predominantly in two of the three Cartesian directions. When confinement is introduced in the z direction for a (001) interface, an xy subband is lowest energy and disperses fairly uniformly in the two-dimensional plane, while xz and yz subbands are higher in energy and approximately one dimensional.

The reduction of the kinetic energy of the xz and yz subbands suggests we consider them for possible ferromagnetic polarization. We therefore adopt a minimal model with three, two-dimensional subbands for the (001) interface, with the Hamiltonian $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_I$, where the kinetic energy is

$$\mathcal{H}_0 = \sum_{\mathbf{k},\alpha} \frac{k_x^2 + k_y^2}{2m_0} d_{0\alpha}^\dagger(\mathbf{k}) d_{0\alpha}(\mathbf{k}) + \sum_{\mathbf{k},\alpha,i=x,y} \left(\Delta + \frac{k_i^2}{2m_i}\right) d_{i\alpha}^\dagger(\mathbf{k}) d_{i\alpha}(\mathbf{k}). \quad (1)$$

Here $d_{0\alpha}$, $d_{x\alpha}$, $d_{y\alpha}$ describe the xy, xz, and yz bands (with spin polarization $\alpha = \uparrow, \downarrow$), respectively, m_i is an effective mass, and Δ is the subband crystal field splitting. We have assumed a tetragonal crystal field symmetry for the (001) interface in Eq. (1), so $m_x = m_y$. Since we always consider the band bottom, this is equivalent to taking a tight-binding model with hopping amplitude $t_i = 1/(2m_i a^2)$, where a is the lattice spacing. Here we make an approximation $t_0 \approx t_x = t_y \equiv t$ so that all the effective masses are equal. We take on-site interactions, of the form

$$\mathcal{H}_I = U \sum_{\mathbf{r},i} n_{i\uparrow}(\mathbf{r}) n_{i\downarrow}(\mathbf{r}) + U' \sum_{\mathbf{r},i\neq j} n_i(\mathbf{r}) n_j(\mathbf{r}) - J_H \sum_{\mathbf{r},i\neq j} \mathbf{S}_i(\mathbf{r}) \cdot \mathbf{S}_j(\mathbf{r}), \quad (2)$$

where $n_{i\alpha}(\mathbf{r}) = d_{i\alpha}^\dagger(\mathbf{r}) d_{i\alpha}(\mathbf{r})$, $n_i(\mathbf{r}) = \sum_\alpha n_{i\alpha}(\mathbf{r})$, and $\mathbf{S}_i(\mathbf{r}) = 1/2 \sum_{\alpha\beta} d_{i\alpha}^\dagger(\mathbf{r}) \boldsymbol{\sigma}_{\alpha\beta} d_{i\beta}(\mathbf{r})$. As usual, we expect the intraorbital interaction Hubbard U to be the largest interaction, with the interorbital interaction U' and the Hund's coupling J_H rather smaller, U'/U, $J_H/U \lesssim 0.3$. Other interactions are typically at least an order of magnitude smaller in three-dimensional transition metal compounds, and interactions between different Ti sites are strongly screened. To proceed, we first project $\mathcal{H}_I$ onto the two-dimensional subbands, which replaces the couplings by renormalized reduced ones, $U \to U/z$, $U' \to U'/z$, $J_H \to J_H/z$, where z is roughly the number of STO unit cells over which the subbands are spread (strictly speaking these factors depend on the bands involved, but below interactions play a key role only for the xz/yz subbands — for which $z \geq 4$ obtains based on subband modeling). We henceforth absorb this renormalization into the couplings.

To analyze the effect of interactions, we now treat the U' and J_H as small (which they are, relative to U) and consider possible ferromagnetic instabilities they induce. With $U' = J_H = 0$, the Hamiltonian is decoupled to three single-band problems, and hence does not support ferromagnetism. However, crucially, the xz and yz subsystems become extremely susceptible to ferromagnetism when they contain a low density of electrons. What we require is the free energy of each orbital subsystem as a function of its magnetization, including the effects of strong on-site U. For the xy subband, which is two-dimensional, an on-site interaction U has little effect, and in the low density (per lattice site) limit studied here, the interactions can be exactly treated by a standard T-matrix ladder summation. The result is simply a Fermi liquid with small Landau parameters, which can be neglected at the level of the present consideration. *A posteriori*, it is justified to assume the magnetization M_0 of the xy subband is small, so that we can just approximate its free energy by the quadratic form $M_0^2/(2\chi_{2d})$, where χ_{2d} is the susceptibility for such a two-dimensional system. Neglecting the Fermi liquid correction, this is $\chi_{2d} = ma^2/(2\pi) = 1/(4\pi t)$.

For the xz and yz subbands, however, due to their one-dimensionality, the situation is radically different. Remarkably, it is known that the susceptibility, χ_{1d}, of a one-dimensional electron gas (1DEG) is highly divergent at low density [18,19]. In particular, it actually diverges as $\chi_{1d} \sim 1/(W x_1^2)$, where W is an energy scale and x_1 is the occupation per site, for any nonzero U. This is a strong interaction effect: the ratio of the interacting to free fermion susceptibility $\chi_{1d}/\chi_{ff} \to \infty$ diverges for $x_1 \to 0$.

We can explain the enhanced susceptibility, and even obtain a general result for the free energy versus magnetization, starting from the fact that low energy scattering is enhanced in one dimension. In particular, for an arbitrary repulsive interaction, the reflection probability for a pair of scattering particles approaches unity when their energy

approaches its minimum; this is true only in one dimension. Consequently, the electrons in a low density 1DEG are almost unable to exchange, and the energy of the ground state becomes almost independent of spin, and equal to that of spinless fermions. In fact, there is a parametrically weak residual exchange coupling, which occurs due to the small transmission probability of colliding electrons. Because the charge degrees of freedom are well ordered on this exchange scale, the spin dependence of the ground state energy is exactly that of a one-dimensional Heisenberg antiferromagnetic chain with an effective exchange interaction $J_{\rm eff}$ much smaller than the one-dimensional Fermi energy ϵ_F, and one "site" per electron in the 1DEG. We expect $J_{\rm eff}/\epsilon_F$ to vanish as $x_1 \to 0$, and since $\epsilon_F \sim t x_1^2$, we guess $J_{\rm eff} \sim W x_1^3$. We have checked that this agrees with all known exact results for the susceptibility of the one-dimensional Hubbard model [18,19]. In the large U limit, we obtain $W \sim 2\pi^2 t^2/(3U)$, while for small U, $W \sim U$. Using the former estimate, we obtain $W \approx (1\text{–}2)t$ for the titanates.

Consequently, we can obtain the free energy for an arbitrary magnetization of the one-dimensional xz and yz subbands. If the spin (per site) in the xy band is M_0 and that in the xz and yz bands is M_1, it is (per site)

$$F = \frac{M_0^2}{2\chi_{2d}} + 2x_1 J_{\rm eff} F_1\left[\frac{M_1}{x_1}, \frac{k_B T}{J_{\rm eff}}\right] - J_H(2M_0M_1 + M_1^2), \tag{3}$$

where $F_1[m, t]$ is the free energy per site of the one-dimensional antiferromagnet chain unit exchange with magnetization m and temperature t. This assumes $x_1 \ll 1$. The first two terms represent the exact thermodynamics for the decoupled orbital subsystems and fully incorporate all the effects of U. The last term is simply the leading first-order term in the expansion of the energy of these states in J_H, presumed small. Equation (3) may also be interpreted in terms of a mean-field treatment of the Hund's coupling only. This is quite analogous to "chain mean-field theory," which has been successfully applied to explain numerous experiments in low-dimensional magnetic materials [20] and is known to be usually quantitatively rather accurate.

Using $J_{\rm eff} = W x_1^3$ and $M_1 \le x_1/2$, we see from Eq. (3) that when $x_1 \lesssim \sqrt{J_H/W}$, the Hund's energy overwhelms the one-dimensional exchange and favors a ferromagnetic state with $M_1 \neq 0$. Remarkably, this occurs for arbitrarily weak Hund's coupling J_H, provided the filling of the upper xz and yz subbands is sufficiently small and, of course, nonzero. This gives a mechanism for ferromagnetism at intermediate carrier density, when the total density is near the critical value needed to just populate the xz and yz subbands, with magnetism disappearing both for smaller and larger carrier density. A quantitative minimization of Eq. (3) is possible since $F_1[m, t]$ is known exactly from the thermodynamic Bethe ansatz [21]. Assuming $J_H \ll t$, W and $x_1 \ll 1$, we obtain a dome-shaped region of ferromagnetism, as shown in Fig. 1. Note that the characteristic maximum temperature scale is of order of $k_B T_c \sim 0.05\sqrt{J_H^3/W}$. From this minimization, we can also obtain the magnetization at all temperatures and fields. In particular we find that at $T = 0$, the xz/yz bands are fully polarized.

It may appear that the one dimensional physics of the above picture is overly exotic and restrictive. However, this is not the case, and can persist up to some reasonable value of t'. This hopping causes a crossover from one-dimensional behavior to two-dimensional Fermi liquid behavior at low energy. By continuity, for small t'/t, this Fermi liquid must have an enhanced spin susceptibility captured by a large Fermi liquid correction F_0^a. However, the eventual two-dimensionality induced by nonzero t' controls the maximum susceptibility achieved at small x_1, and if this effect is too large, the ferromagnetic instability may be entirely removed. The susceptibility divergence is cut off when the distance between the Fermi energy and the bottom of the xz and yz bands is comparable to the hopping t', i.e., $t x_1^2 \sim t'$. The same condition describes the change from an open Fermi surface to an elliptical one. This gives the condition $t' \lesssim J_H t/W$ for the ferromagnetic phase to occur (we neglect numerical prefactors here due to the imprecision of the matching argument). Of course, when t' is substantial, the magnitude of the magnetization and of T_c will be reduced from the one-dimensional values given in Fig. 1, further increasing the tendency to low T_c and small net moment.

Recently the (110) and (111) LAO-STO interfaces have also been prepared experimentally [22,23], and both interfaces appear to support STO electron gases, though the (111) interface is polar and the (110) is not. As listed in Table I, these two interfaces have different local crystal field environments and hence different local orbital configuration from the (001) interface. Can these two interfaces also support ferromagnetism—under ideal disorder-free conditions—at certain electron fillings?

As usual, the e_g doublets are always higher in energy and do not play any role. For the (110) interface, the three t_{2g} orbitals are split into three nondegenerate orbitals:

TABLE I. The relevant local crystal symmetries and local orbital states for different interfaces. ";" delimits the sets of locally degenerate orbital states. Note that there could be a small hybridization between $(1/\sqrt{2})(xz + yz)$ and xy orbitals for the (110) interface.

Interfaces	Symmetry	Local orbitals
(001)	Fourfold rotation	xz, yz; xy
(110)	Twofold rotation	$(1/\sqrt{2})(xz + yz)$; xy; $(1/\sqrt{2})(xz - yz)$
(111)	Threefold rotation	$(1/\sqrt{3})(xy + e^{i(2\pi/3)}yz + e^{-i(2\pi/3)}xz)$, $(1/\sqrt{3})(xy + e^{-i(2\pi/3)}yz + e^{i(2\pi/3)}xz)$; $(1/\sqrt{3})(xy + yz + xz)$

$(1/\sqrt{2})(xz + yz)$, $(1/\sqrt{2})(xz - yz)$, and xy. In the first approximation, the local hybridization between $(1/\sqrt{2}) \times (xz + yz)$ and xy orbitals may be neglected. When these two orbitals form bands, they are also quasi-one-dimensional, just like xz and yz orbitals for the (001) interface. Hopping among $\frac{1}{\sqrt{2}}(xz + yz)$ $(/xy)$ orbitals occurs most strongly with neighbors along z $(/[1\bar{1}0])$ lattice directions. The $(1/\sqrt{2})(xz - yz)$ subband is two dimensional and its band bottom is the lowest among the three subbands. Due to the reduced symmetry of the (110) interface, the two upper quasi-one-dimensional subbands are split. Based on our above discussion of ferromagnetic instability for the (001) interface, we also expect emergent ferromagnetism for the (110) interface when the filling of the quasi-one-dimensional subband is sufficiently small. Because the two upper quasi-one-dimensional subbands are not degenerate, there may even exist two ferromagnetic regimes as the electron filling of the two upper subbands is increased. One should note that the discussion here assumes no hybridization between $(1/\sqrt{2})(xz + yz)$ and xy orbitals. In reality, there is always small hybridization between these two orbitals. If this hybridization is very small (smaller than $\mathcal{O}(\sqrt{J_H/J})$), the resulting two-dimensional Fermi liquid should still have a large spin susceptibility and ferromagnetism can still be present.

For the (111) interface, although locally the crystal field splits three t_{2g} orbitals into one a_{1g} state, $(1/\sqrt{3})(xy + yz + xz)$, and two e'_{2g} states, $(1/\sqrt{3})(xy + e^{i(2\pi/3)}yz + e^{-i(2\pi/3)}xz)$ and $(1/\sqrt{3})(xy + e^{-i(2\pi/3)}yz + e^{i(2\pi/3)}xz)$, the electron hopping strongly hybridizes three orbitals and leads to two-dimensional Fermi liquids. Hence, no ferromagnetism arises in this case.

In contrast to the itinerant mechanism discussed here, other theoretical works have instead proposed mechanisms relying on localized electron moments. While we believe that *Mott* localization of electrons near the interface should not occur for ideal structures, sufficient disorder and interactions together might create some truly localized moments. If the localized electron mechanisms are correct, we predict significant dependence of the ferromagnetism on disorder, and indeed that it should weaken as sample quality is improved. The itinerant mechanism discussed here has its own distinct predictions, e.g., Fig. 1, and the fact that the polarization resides in xz/yz bands, which may be tested by x-ray dichroism experiments. Further, varying the electron concentration away from the critical density by tuning the back gate voltage may easily suppress the ferromagnetism, and ferromagnetism should be absent at the (111) interface; neither prediction applies for the local moment mechanism [24].

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