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Tetrahis(dimethylamino)ethylene-C_{60}: Multicomponent superexchange and Mott ferromagnetism

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An interacting, quasi-one-dimensional, orbitally degenerate hopping model is introduced, motivated by the structure of tetrahis(dimethylamino)ethylene (TDAE)-C_{60}. The ground state is a Mott insulator for average ionization ${C_{60}}^-$, with several possible magnetically ordered phases. The effective Hamiltonian is derived via a multicomponent superexchange transformation. Orbital degeneracy is represented as an additional "isospin" degree of freedom. We explore the phase diagram of the one-dimensional model in interaction parameter space, and identify various special points: (i) an SU(4) antiferromagnet (solved by Sutherland and approximated by a large-N mean-field theory), (ii) a spin-ferromagnet/isospin-antiferromagnet, and (iii) an isospin-ferromagnet/spin-antiferromagnet. The exact ground states for (ii) and (iii) are proven using Marshall's theorem. Away from these points we determine the phase diagram in the classical limit. Three-dimensional ordering for case (ii) is found using interchain mean-field theory. We find two transition temperatures: $T_C \propto \sqrt{J_\perp J_\parallel}$ for the ferromagnetic spin order and $T_N \propto J_\perp$ for the antiferromagnetic isospin ordering, where $J_\parallel$, $J_\perp$ are the intrachain and interchain couplings, respectively. We discuss the relevance of our theory to recent experiments on TDAE-C_{60}.

I. INTRODUCTION

The synthesis of the fullerene (C_{60}) into molecular crystals with electron donors has resulted in materials with remarkable electronic properties. First, superconductivity was discovered in alkali fullerenes A_3C_{60} (A = K, Cs, Rb) (Ref. 1) at relatively high temperatures of $T_c \leq 33$ K.[2] Soon thereafter ferromagnetism was found in TDAE-C_{60} at $T_c \approx 16$ K,[3,4] where TDAE is tetrakis(dimethylamino)ethylene, $C_2N_4(CH_3)_8$ (the stoichiometry is a 1:1 ratio of TDAE to C_{60}). Some striking aspects of this material are its value of T_c—relatively large for an organic ferromagnet—and its nonmetallic conductivity, suggestive of Mott-Hubbard localization.[5] This is perhaps unusual, since superexchange naively predicts antiferromagnetic interactions between localized spins in many such systems.

An obvious question is raised: *Are superconductivity in A_3C_{60} and ferromagnetism in TDAE-C_{60} related?* While superconductivity involves effectively attractive interactions, magnetism is usually believed to result from repulsive Coulomb forces. (A similar mystery underlies the proximity of high-T_c superconductivity to antiferromagnetism in the high-T_c cuprates.) The similarity between the materials is that both involve partially filled conduction bands made of t_{1u} orbitals of C_{60}. Also one expects similar intramolecular electron-vibron and electron-electron interactions.

The primary differences between the two systems are in their crystalline symmetries and C_{60} ionizations. While A_3C_{60} is a fcc crystal with cubic symmetry in which the C_{60} molecules are triply ionized (${C_{60}}^{3-}$), TDAE-C_{60} has a c-centered monoclinic unit cell,[6] which gives rise to preferred hopping along the c axis, and singly ionized ${C_{60}}^-$. One of our aims is to show here that these differences are indeed responsible for the difference between the superconductors and the ferromagnet. Our model elaborates upon the physical mechanism identified by Seshadri *et al.*,[7] who pointed out that ferromagnetism is a consequence of both molecular degeneracies and a triplet ${C_{60}}^{2-}$ ground state.[7a] We also investigate the effects of hopping and interactions in an anisotropic crystal environment. The hopping Hamiltonian is mapped, through superexchange, to a multicomponent spin model which possesses both true spin as well as "isospin" degrees of freedom, the latter representing doubly (rather than triply) degenerate molecular orbitals in the presence of a symmetry-breaking crystal field.

The inclusion of crystal fields, we show, can give rise to insulating ferromagnetic behavior even when there is pair binding in the isolated C_{60}-${C_{60}}^{2-}$ system.

In the quasi-one dimensional limit, we identify special points where the ground state is exactly known. For degenerate pseudopotentials we obtain an SU(4) exchange Hamiltonian in the fundamental representation, which has been solved by Sutherland using Bethe's ansatz.[8,9] In another limit (large singlet-triplet splitting), we prove by a generalized Marshall theorem that the ground state is the fully polarized Heisenberg ferromagnet. To our knowledge, this is a rare example of proven local ferromagnetism in a microscopically based model of electron interactions. (Other examples are the Nagaoka ferromagnet of one hole in the infinite-U Hubbard model and the Hubbard model on a bipartite lattice with unequal sublattice sizes. Neither is easy to realize physically.[9a]) The global phase diagram and the effect of interchain coupling will be calculated within a mean-field approximation.

II. HOPPING MODEL

Electron-spin-resonance measurements[10] confirm that TDAE donates a single electron to C_{60}. Thus we consid-

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er a conduction band formed by a tight-binding hopping on a lattice of C_{60} molecules with a filling of one electron per site. In Ref. 6, the intermolecular distances were found to be 9.98 Å along the c axis and 10.25 Å within the a-b plane. (In comparison, in A_3C_{60}, the distances range from 10.07 to 10.25 Å.)

The composition rules of $t_{1u} \otimes t_{1u} = a_g \oplus h_g \oplus t_{1g}$ for representations of the icosahedral group (the symmetry group of C_{60}) are identical to the corresponding representations of the spherical group. We simplify notations by using angular momentum quantum numbers instead of the icosahedral group representations for the molecular orbitals:

$$\begin{aligned} &\text{icosahedral} \rightarrow \text{spherical} \, , \\ &|a_g\rangle \rightarrow |0,0\rangle \, , \\ &|t_{1g}\rangle \rightarrow |1,m_L\rangle \quad (m_L = -1,0,1) \, , \\ &|h_g\rangle \rightarrow |2,m_L\rangle \quad (m_L = -2,-1,0,1,2) \, , \end{aligned} \tag{1}$$

where $|L,m_L\rangle$ are eigenstates of angular momentum operators $\mathbf{L}^2$ and L^z. We identify the z axis with the c direction.

We denote by $t_{ll'}(\boldsymbol{\delta})$ the hopping matrix element between t_{1u} orbitals l and l' on sites separated by a nearest-neighbor distance δ. The Hamiltonian is then given by

$$\begin{aligned} \mathcal{H} = &-\frac{1}{2} \sum_{\substack{\mathbf{i},\boldsymbol{\delta},\sigma \\ l,l'}} [t_{ll'}(\boldsymbol{\delta}) c^\dagger_{l\sigma}(\mathbf{i}) c_{l'\sigma}(\mathbf{i}+\boldsymbol{\delta}) + \text{H.c.}] \\ &+ \sum_{\mathbf{i}} \mathcal{H}_{\text{ion}}(\mathbf{i}) + \sum_{\mathbf{i}} \mathcal{H}_{\text{CF}}(\mathbf{i}) \, , \end{aligned} \tag{2}$$

where $c^\dagger_{l\sigma}(\mathbf{i})$ creates, at site $\mathbf{i}$, an electron of spin polarization $\sigma = \uparrow, \downarrow$ and orbital quantum number $l = -1,0,1$ (the t_{1u} triplet). $\mathcal{H}_{\text{ion}}(i)$ is the ionic interaction Hamiltonian which discourages multiple-electron occupancy of the C_{60} molecule at $\mathbf{i}$, and $\mathcal{H}_{\text{CF}}(\mathbf{i})$ is the crystal-field Hamiltonian discussed below, which partially lifts the threefold orbital degeneracy of the t_{1u} level.

To reflect the quasi-one-dimensionality of TDAE-C_{60},[6] we assume that the largest hopping integrals arise when $\boldsymbol{\delta}$ lies along the c axis.

$\mathcal{H}_{\text{ion}}$ is defined by the two-electron spectrum on a C_{60} molecule. In the absence of crystal fields, the two-electron states in the t_{1u} shell form three multiplets,

$$|L,S;m_L,m_S\rangle = \begin{cases} |2,0;m_L,0\rangle \times 5 \, , \\ |1,1;m_L,m_S\rangle \times 9 \, , \\ |0,0;0,0\rangle \times 1 \, , \end{cases} \tag{3}$$

where L, S, m_L, and m_S denote the total angular momentum, total spin, angular momentum polarization, and spin polarization quantum numbers, respectively. In second-quantized notation, these states are written as

$$|L,S;m_L,m_S\rangle \equiv \sum_{\alpha\beta} M_{\alpha,\beta}(L,S;m_L,m_S) c^\dagger_\alpha c^\dagger_\beta |0\rangle \, , \tag{4}$$

where $\alpha = \{l,\sigma\}$ ranges over six possible values. The above equation defines the matrices $M_{\alpha\beta}(\Lambda)$, where $|\Lambda\rangle$ is one of the eigenstates of $\mathcal{H}_{\text{ion}}$. The electron-electron and the electron-vibron interactions respect the molecular symmetry and are independent of spin and therefore do not lift the degeneracies given in Eq. (3). These intramolecular interactions are parametrized by three pseudopotentials u_0, u_1, and u_2, which are the interaction energies in the $L=0$, $L=1$, and $L=2$ angular momentum channels, respectively. The u_L are the low-frequency limit of the two-electron vertex function, which can be computed by taking differences of *relaxed* ground-state energies E^n of the $C_{60}{}^{n-}$ ions:[11,12]

$$u_L = E^2_L - 2E^1 + E^0 \, , \tag{5}$$

where L is the total angular momentum of the two-electron states. (The spin S of the two-electron state with angular momentum L is determined from the requirement that $L+S$ be even.) Attractive pseudopotentials $u_L < 0$ are also called "pair binding energies".[12]

Thus we can write $\mathcal{H}_{\text{ion}}$ as

$$\mathcal{H}_{\text{ion}} = \sum_{\substack{L,S \\ m_L,m_S}} u_L |L,S;m_L,m_S\rangle\langle L,S;m_L,m_S| \, . \tag{6}$$

III. CRYSTAL-FIELD EFFECT

The crystal-field interaction in the t_{1u} shell is modeled by

$$\mathcal{H}_{\text{CF}} = -\Delta \sum_{\mathbf{i},l,\sigma} l^2 c^\dagger_{l\sigma}(\mathbf{i}) c_{l\sigma}(\mathbf{i}) \, , \tag{7}$$

where $\Delta > 0$. Equation (7) implicitly assumes that the crystal fields resolve the threefold t_{1u} degeneracy into a doublet ($l = \pm 1$) at energy $-\Delta$ and a singlet ($l=0$) at zero energy. In fact, this is true only for crystal fields which are rotationally symmetric about an axis which pierces the center of a pentagonal face of C_{60}. In general, a tetragonal or monoclinic crystalline symmetry will resolve the triply degenerate t_{1u} level into *three* distinct levels. We shall show in the following that ferromagnetism in TDAE-C_{60} follows from the (twofold) degeneracy of molecular orbitals in the conduction band. If one adopts the model of Eq. (7), then $\Delta < 0$ would yield a nondegenerate conduction band and hence antiferromagnetic superexchange, a state of affairs apparently ruled out by experiment.

We also assume $\Delta/|u_L| \gg 1$, which allows us to ignore the higher-lying $l=0$ orbital entirely, restricting our attention to the $l = \pm 1$ states. Hopping along the chains is then assumed to preserve the orbital quantum number l. This, too, is a rather strong assumption and one which cannot be justified given our lack of information concerning the relative orientation of consecutive C_{60} molecules along the c axis. It seems likely to us that the essential features of our model (e.g., ferromagnetism) will not be severely altered by retaining all three of the t_{1u} orbitals in a more complicated Hamiltonian, with a weakly split low-lying doublet.

We are then led to the Hamiltonian

$$\mathcal{H}=\mathcal{H}^{\parallel}_{\rm hop}+\mathcal{H}^{\perp}_{\rm hop}+\mathcal{H}_{\rm ion}\ ,$$

$$\mathcal{H}^{\parallel}_{\rm hop}=-t^{\parallel}\sum_{\mathbf{i},l,\sigma}[c^{\dagger}_{l\sigma}(\mathbf{i})c_{l\sigma}(\mathbf{i}+\mathbf{c})+\text{H.c.}]\ ,$$

$$\mathcal{H}^{\perp}_{\rm hop}=-\frac{1}{2}\sum_{\substack{\mathbf{i},\boldsymbol{\delta}_\perp\\ l,l',\sigma}}t^{\perp}_{ll'}(\boldsymbol{\delta}_\perp)[c^{\dagger}_{l\sigma}(\mathbf{i})c_{l'\sigma}(\mathbf{i}+\boldsymbol{\delta}_\perp)+\text{H.c.}]\ ,$$

$$\mathcal{H}_{\rm ion}=\sum_{\mathbf{i},\Lambda}\bar{u}_{\Lambda}|\Lambda(\mathbf{i})\rangle\langle\Lambda(\mathbf{i})|\ , \tag{8}$$

where the two-electron states $|\Lambda\rangle$ are those of Eq. (3) after projection onto the subspace containing no $l=0$ orbitals. This projection eliminates six of the nine $L=1$, $S=1$ states and three of the five $L=2$, $S=0$ states. In addition, another state from the $L=2$, $S=0$ multiplet, as well as the $L=0$, $S=0$ states, yields the same result under projection (a state of indefinite L).

Explicitly, the surviving states $|\Lambda\rangle$ are a triplet of energy $\bar{u}_1\equiv u_1$,

$$c^{\dagger}_{+\uparrow}c^{\dagger}_{-\uparrow}|0\rangle=|1,1;0,1\rangle\ ,$$

$$\frac{1}{\sqrt{2}}(c^{\dagger}_{+\uparrow}c^{\dagger}_{-\downarrow}+c^{\dagger}_{+\downarrow}c^{\dagger}_{-\uparrow})|0\rangle=|1,1;0,0\rangle\ , \tag{9}$$

$$c^{\dagger}_{+\downarrow}c^{\dagger}_{-\downarrow}|0\rangle=|1,1;0,-1\rangle\ , \tag{10}$$

a doublet of energy $\bar{u}_2\equiv u_2$,

$$c^{\dagger}_{+\uparrow}c^{\dagger}_{+\downarrow}|0\rangle=|2,0;2,0\rangle\ ,$$

$$c^{\dagger}_{-\uparrow}c^{\dagger}_{-\downarrow}|0\rangle=|2,0;-2,0\rangle\ , \tag{11}$$

and a singlet of energy $\bar{u}_0\equiv\frac{2}{3}u_0+\frac{1}{3}u_2$,

$$\frac{1}{\sqrt{2}}(c^{\dagger}_{+\uparrow}c^{\dagger}_{-\downarrow}-c^{\dagger}_{+\downarrow}c^{\dagger}_{-\uparrow})|0\rangle$$

$$=\frac{1}{\sqrt{3}}|2,0;0,0\rangle-\frac{\sqrt{2}}{\sqrt{3}}|0,0;0,0\rangle\ . \tag{12}$$

These states define the matrices $M^{\Lambda}_{\alpha\beta}$ through

$$|\Lambda\rangle\equiv\frac{1}{2}\sum_{\alpha\beta}M^{\Lambda}_{\alpha\beta}c^{\dagger}_{\alpha}c^{\dagger}_{\beta}|0\rangle\ . \tag{13}$$

Calculations of the pseudopotentials u_L, based on microscopic models of electron-electron and electron-vibron interactions, have appeared in the recent literature.[13,14] For an order-of-magnitude estimate of the pair binding energy u_0 of $C_{60}{}^{0-}$-$C_{60}{}^{2-}$ in K_3C_{60}, we appeal to the weak-coupling BCS formula

$$T_c=\bar{\omega}\exp\left[-\frac{1}{N(\epsilon_F)|u_0|}\right] \tag{14}$$

and the experimental values[15] of $T_c=19$ K and $\bar{\omega}=1100$ cm^{-1}, together with band theory estimates[16] of the density of states $N(\epsilon_F)=7.2$ states/eV spin. This yields

$$u_0(A_3C_{60})\approx-0.03\ \text{eV}\ . \tag{15}$$

This energy is a sum of the negative electron-vibron interactions[17,18] and positive (or negative[12]) electron-electron interactions. However, since $\bar{u}_0=\frac{2}{3}u_0+\frac{1}{3}u_2$, we have that $\bar{u}_0>0$ if $u_2>2|u_0|$, thus explaining why TDAE-C_{60} is not a superconductor, even if $u_0<0$ is pair binding.

For the hopping matrix $t^{\perp}_{ll'}$ $(\boldsymbol{\delta}_\perp)$, we assume the simple form

$$t^{\perp}_{ll'}(\boldsymbol{\delta}_\perp)=\begin{bmatrix}t^{\perp}_1 & \pm t^{\perp}_2\\ \pm t^{\perp}_2 & t^{\perp}_1\end{bmatrix}\ , \tag{16}$$

where the sign of the off-diagonal elements is positive for $\boldsymbol{\delta}_\perp=\pm\hat{\mathbf{x}}$ and negative for $\boldsymbol{\delta}_\perp=\pm\hat{\mathbf{y}}$.

IV. MULTICOMPONENT SUPEREXCHANGE HAMILTONIAN

Experiments have shown that TDAE-C_{60} is insulating at low temperatures, consistent with the existence of a gap $(\bar{u})$ to charge fluctuations. We believe this material to be a Mott insulator, and in this section we derive an effective Hamiltonian for the low-lying spin and orbital excitations. The superexchange model is an expansion in the small parameter $t^{\parallel}/\bar{u}$, though presumably even for $t^{\parallel}/\bar{u}\gtrsim 1$ a similar superexchange model with renormalized couplings may be applied. Within the subspace of singly occupied sites, one has the superexchange Hamiltonian

$$\tilde{\mathcal{H}}=-P\mathcal{H}_{\rm hop}(1-P)(\mathcal{H}_{\rm ion})^{-1}(1-P)\mathcal{H}_{\rm hop}P\ , \tag{17}$$

where P projects onto the subspace where each site is singly occupied. For a particular link $(\mathbf{ij})$, one has

$$\tilde{\mathcal{H}}(\mathbf{ij})=\sum_{\substack{\alpha,\beta\\ \bar{\alpha},\bar{\beta}}}J^{\bar{\alpha}\beta}_{\alpha\bar{\beta}}S^{\alpha}_{\beta}(\mathbf{i})S^{\bar{\beta}}_{\bar{\alpha}}(\mathbf{j})+(\mathbf{i}\leftrightarrow\mathbf{j})\ ,$$

$$J^{\bar{\alpha}\beta}_{\alpha\bar{\beta}}=\sum_{\Lambda}\frac{1}{\bar{u}_{\Lambda}}(tM^{\Lambda})_{\alpha\bar{\beta}}(M^{\Lambda}t)_{\bar{\alpha}\beta}\ , \tag{18}$$

where

$$S^{\alpha}_{\beta}(\mathbf{i})=c^{\dagger}_{\alpha}(\mathbf{i})c_{\beta}(\mathbf{i}) \tag{19}$$

is a generalized spin operator at site $\mathbf{i}$ and tM^{Λ} is the matrix product of t and M^{Λ}. The indices α, β, $\bar{\alpha}$, and $\bar{\beta}$ run over the four single-particle orbitals $\{+\uparrow,+\downarrow,-\uparrow,-\downarrow\}$, and single occupancy means that $\text{Tr}S(\mathbf{i})=1$. Hence there are 15 independent components to $S^{\alpha}_{\beta}(\mathbf{i})$, corresponding to the 15 generators of the group SU(4). The four possible states at each site constitute a basis in the fundamental representation of SU(4).

It is also possible to represent the S^{α}_{β} in terms of spin and "isospin" operators, viz.,

$$S^{\mu}_{\mathbf{i}}=\frac{1}{2}\sum_{l,\sigma,\sigma'}c^{\dagger}_{l\sigma}(\mathbf{i})\tau^{\mu}_{\sigma\sigma'}c_{l\sigma'}(\mathbf{i})\ ,$$

$$I^{\nu}_{\mathbf{i}}=\frac{1}{2}\sum_{l,l',\sigma}c^{\dagger}_{l\sigma}(\mathbf{i})\tau^{\nu}_{ll'}c_{l'\sigma}(\mathbf{i})\ , \tag{20}$$

where τ are the Pauli matrices. The 15 independent elements of S^{α}_{β} can then be expressed in terms of the 15 operators $\{S^{\mu},I^{\nu},S^{\mu}I^{\nu}\}$.

V. ONE-DIMENSIONAL LIMIT

We first consider the purely one-dimensional (1D) case, where $t^{\perp}_{ll'}=0$. In this limit, the effective Hamiltonian $\tilde{\mathcal{H}}$ is parametrized by three superexchange constants

$$J_M \equiv \frac{2(t^{\parallel})^2}{\bar{u}_M} , \quad M=0,1,2 , \tag{21}$$

and we can write

$$\begin{aligned}\tilde{\mathcal{H}}=\sum_n (A\mathbf{S}_n\cdot\mathbf{S}_{n+1}+B\mathbf{I}_n\cdot\mathbf{I}_{n+1} \\ +CI^z_nI^z_{n+1}+D\mathbf{S}_n\cdot\mathbf{S}_{n+1}\mathbf{I}_n\cdot\mathbf{I}_{n+1} \\ +E\mathbf{S}_n\cdot\mathbf{S}_{n+1}I^z_nI^z_{n+1}+F) ,\end{aligned} \tag{22}$$

where

$$\begin{aligned} A&=-\tfrac{1}{2}J_1+J_2+\tfrac{1}{2}J_0 , \\ B&=\tfrac{3}{2}J_1-\tfrac{1}{2}J_0 , \\ C&=J_0-J_2 , \\ D&=2J_1+2J_0 , \\ E&=4J_2-4J_0 , \\ F&=-\tfrac{3}{8}J_1-\tfrac{1}{4}J_2-\tfrac{1}{8}J_0 . \end{aligned} \tag{23}$$

This model possesses a global SU(2)×U(1) symmetry; i.e., $\tilde{\mathcal{H}}$ commutes with $\sum_n\mathbf{S}_n$ and with $\sum_n I^z_n$. Enlarged symmetries occur when $J_0=J_2$, where the symmetry group is SU(2)×SU(2), and when $J_0=J_1=J_2$, where the symmetry group is SU(4).

A. SU(4) point

In the limit where $\bar{u}_1=\bar{u}_2=\bar{u}_0\equiv\bar{u}$ and hence

$$J^{\bar{\alpha}\beta}_{\alpha\bar{\beta}}=\frac{2(t^{\parallel})^2}{\bar{u}}\delta_{\alpha\bar{\alpha}}\delta_{\beta\bar{\beta}} , \tag{24}$$

the Hamiltonian of Eq. (18) possesses a global SU(4) symmetry and is given by a sum over noninteracting spin chains, each of which is governed by

$$\mathcal{H}=J\sum_n\sum_{\alpha,\beta}S^{\alpha}_{\beta}(n)S^{\beta}_{\alpha}(n+1) , \tag{25}$$

where $J=2(t^{\parallel})^2/\bar{u}$. While this model is not directly relevant to TDAE-C_{60} (it is not ferromagnetic in the spin channel), its identification on our phase diagram is worthwhile because it is exactly solvable. The model has been solved using Bethe's ansatz by Sutherland,[9] who solved the problem of the general SU(P) Heisenberg antiferromagnet in the fundamental representation, the $P=2$ version being the one originally solved by Bethe himself. The general model features $P-1$ gapless elementary excitation branches. We presume, based on what happens in the SU(2) model,[19] that for a chain of N sites, where N is an integer multiple of P, the ground state is an SU(P) singlet and the low-lying excitations transform according either to the singlet or adjoint representation.

This is essentially what happens in the fermion mean-field theory of the SU(P) antiferromagnet.[20–22] The mean-field Hamiltonian is given by

$$\begin{aligned}\mathcal{H}^{\mathrm{MF}}=\sum_{k,\alpha}(\lambda+2Q\cos k)c^{\dagger}_{\alpha}(k)c_{\alpha}(k) \\ +2\left[\frac{Q^2}{J}-\lambda\right]N ,\end{aligned} \tag{26}$$

and minimization of the ground-state energy for the $P=4$ case gives $Q=-J/\pi\sqrt{2}$, $\lambda=J/\pi$, and a mean-field dispersion

$$\epsilon(k)=\frac{J}{\pi}(1-\sqrt{2}\cos k) . \tag{27}$$

One thus has four degenerate quarter-filled ($k_F=\frac{1}{4}\pi$) bands for $P=4$. Although there is no true long-ranged order, the susceptibility diverges at the nesting wave vector $2k_F=\frac{1}{2}\pi$, which describes a commensurate spin-density wave of period 4. The period 4 arises because the spin chain is in its fundamental representation, and by "4-ality" one needs four sites to make a singlet. [A separate class of SU(P) models in which sites on alternating sublattices transform according to fundamental and antifundamental[23] allow a singlet to be formed within any consecutive *pair* of sites, and such models exhibit long-range order via dimerization.]

B. Special points of SU(2)×SU(2) symmetry

Along the surface $\bar{u}_2=\bar{u}_0$, our Hamiltonian possesses an SU(2)×SU(2) symmetry. There are then two special limits in which we can determine the exact ground state. One such limit is $\bar{u}_0\rightarrow\infty$, where the interactions are ferromagnetic in the spin channel and antiferromagnetic in the isospin channel; we refer to this as the "$F\times A$ model." The other limit ("$A\times F$ model") occurs when $\bar{u}_1\rightarrow\infty$, where the interactions are antiferromagnetic in the spin channel and ferromagnetic in the isospin channel. The corresponding Hamiltonians are given by

$$\mathcal{H}_{F\times A}=-\frac{4(t^{\parallel})^2}{\bar{u}_1}\sum_n(\mathbf{S}_n\cdot\mathbf{S}_{n+1}+\tfrac{3}{4})(\tfrac{1}{4}-\mathbf{I}_n\cdot\mathbf{I}_{n+1}) , \tag{28}$$

$$\mathcal{H}_{A\times F}=-\frac{4(t^{\parallel})^2}{\bar{u}_0}\sum_n(\tfrac{1}{4}-\mathbf{S}_n\cdot\mathbf{S}_{n+1})(\mathbf{I}_n\cdot\mathbf{I}_{n+1}+\tfrac{3}{4}) . \tag{29}$$

It is possible to prove the following.

Theorem. $\mathcal{H}_{F\times A}$ has a ground state in which the spin variables are fully polarized and the isospin wave function is given by Bethe's solution to the spin-half antiferromagnet, i.e.,

$$|F\times A\rangle=|F\rangle_S\otimes|\mathrm{Bethe}\rangle_I . \tag{30}$$

A corresponding result holds for $\mathcal{H}_{A/F}$, with spin and isospin variables exchanged.

Proof. Because of SU(2)⊗SU(2) symmetry of $\mathcal{H}_{F\times A}$, the total spin S_{tot}, total isospin I_{tot}, and their polarizations along the $\hat{\mathbf{z}}$ axis (M_S and M_I, respectively) are good quantum numbers. We now follow Lieb and Mattis' proof[24] of the Marshall theorem[25] for the Heisenberg model, first performing a π rotation about the $\hat{\mathbf{z}}$ axis of the isospin operators on odd-numbered sites:

$$
\begin{aligned}
I^+_{2l+1} &\to -I^+_{2l+1}\ ,\\
I^-_{2l+1} &\to -I^-_{2l+1}\ ,\\
I^z_{2l+1} &\to I^z_{2l+1}\ ,
\end{aligned}
\tag{31}
$$

which transforms the Hamiltonian into

$$
\begin{aligned}
\mathcal{H}_{F\times A} \to J\sum_n &\left(I^z_n I^z_{n+1} - \tfrac{1}{2} I^+_n I^-_{n+1} - \tfrac{1}{2} I^-_n I^+_{n+1} - \tfrac{1}{4}\right)\\
&\times\left(\mathbf{S}_n\cdot\mathbf{S}_{n+1} + \tfrac{3}{4}\right) \equiv \mathcal{H}'_{F\times A}\ ,
\end{aligned}
\tag{32}
$$

which can be seen to be a nonpositive ("negative semidefinite") operator in the transformed Ising basis

$$
|\{m^S_n\},\ \{m^I_n\}\rangle \to (-1)^{\sum_n (1/2+m^I_n)}|\{m^S_n\},\ \{m^I_n\}\rangle\ . \tag{33}
$$

The accessibility of all states within a given magnetization sector by repeated application of the Hamiltonian implies (see Ref. 24) that the ground state in the sector $(M_S,M_I)=(0,0)$ can be chosen to be positive definite in the sublattice-rotated Ising basis; i.e., it obeys Marshall's sign rule in the untransformed Ising basis

$$
\begin{aligned}
|\Psi_0\rangle = \sum_{\substack{\{m^S_n,m^I_n\}\\ M_S=M_I=0}} &(-1)^{\sum_n (1/2+m^I_n)} f(\{m^S_i,m^I_i\})\\
&\times|\{m^S_i\},\{m^I_i\}\rangle\ ,
\end{aligned}
\tag{34}
$$

where

$$
f(\{m^S_i,m^I_i\})>0\ . \tag{35}
$$

Since the same Marshall signs hold for the state of Eq. (30), which has a representative in the sector $(M_S,M_I)=(0,0)$, the overlap

$$
\langle\Psi_0|F\times A\rangle\neq 0\ , \tag{36}
$$

which proves that the ground state $|\Psi_0\rangle$ must also have the quantum numbers $S_{\text{tot}}=\frac{1}{2}N$, and $I_{\text{tot}}=0$.

By symmetry, the ground state does not depend on M_S, and we are free to choose $M_S=\frac{1}{2}N$ as a representative. Note that $|\Psi_0\rangle$ is indeed an eigenstate of the spin-triplet projection operator $(\mathbf{S}_n\cdot\mathbf{S}_{n+1}+\frac{3}{4})$ with eigenvalue one. It follows from Eq. (28) that the isospin part of the wave function is the ground state of the spin-half antiferromagnetic Heisenberg chain, given by Bethe's ansatz. This completes the proof.

Unfortunately, our theorem does not allow us to determine the excitations of $\mathcal{H}_{F\times A}$ in an exact form. One can construct exact excited states of the Des-Cloizeaux-Pearson Faddeev-Takhtajan type in the isospin channel,[19,26] but the construction of ferromagnetic spin waves is not straightforward because of the antiferromagnetic isospin correlations. Nevertheless, gapless ferromagnetic magnons do exist, since the ground state breaks SU(2) in the spin channel. We can model these modes in the single-mode approximation[27] (SMA) as

$$
\begin{aligned}
|k\rangle &= S^-_k|\Psi_0\rangle/\sqrt{\langle\Psi_0|S^+_{-k}S^-_k|\Psi_0\rangle}\ ,\\
S^\alpha_k &= \frac{1}{\sqrt{N}}\sum_n e^{-ikn}S^\alpha_n\ .
\end{aligned}
\tag{37}
$$

This gives a trial dispersion of

$$
\omega(k)=\langle k|\mathcal{H}_{F\times A}|K\rangle = 2\ln(2)J(1-\cos k)\ , \tag{38}
$$

with $J=2(t^{\parallel})^2/\bar{u}_1$. Thus the role of the antiferromagnetic isospin correlations is to reduce the ferromagnetic magnon bandwidth in the spin channel by a factor of $\langle\frac{1}{4}-\mathbf{I}_n\cdot\mathbf{I}_{n+1}\rangle=\ln 2$.

C. Classical phase diagram

The ground-state phase diagram can be described by the two dimensionless ratios $\bar{u}_0/\bar{u}_1$ and $\bar{u}_2/\bar{u}_1$. We study the classical (large-S, large-I) ground states of Eq. (28) by minimizing the bond energies with respect to $\langle\mathbf{S}_n\cdot\mathbf{S}_{n+1}\rangle$, $\langle\mathbf{I}_n\cdot\mathbf{I}_{n+1}\rangle$, and the ordering direction of $\mathbf{I}$. The results are plotted in Fig. 1. Also identified in the figure are the three special models discussed above. It is interesting to note that the SU(4) symmetry point is at the border of four distinct classical phases. Thus we see that the high ground-state degeneracy of the classical SU(4) point is lifted by quantum fluctuations. Although the ground state of the SU(4)-invariant Hamiltonian of Eq. (25) has no long-range order, it exhibits gapless elementary excitations at wave vectors $k=0,\ \frac{1}{2}\pi,\pi,\frac{3}{2}\pi$.

VI. THREE-DIMENSIONAL ORDERING IN THE $F\times A$ MODEL

Consider an anisotropic Hamiltonian of the form

$$
\begin{aligned}
\mathcal{H}=\mathcal{H}_0 &- \frac{1}{2}\sum_{\mathbf{i},\boldsymbol{\delta}_\perp} J^{ab}_\perp(\boldsymbol{\delta}_\perp)Q_a(\mathbf{i})Q_b(\mathbf{i}+\boldsymbol{\delta}_\perp)\\
&-\sum_{\mathbf{i}} h^a_0(\mathbf{i})Q_a(\mathbf{i})\ ,
\end{aligned}
\tag{39}
$$

where $\mathcal{H}_0$ is a sum over one-dimensional Hamiltonians for a set of chains, $\boldsymbol{\delta}_\perp$ is a nearest-neighbor separation

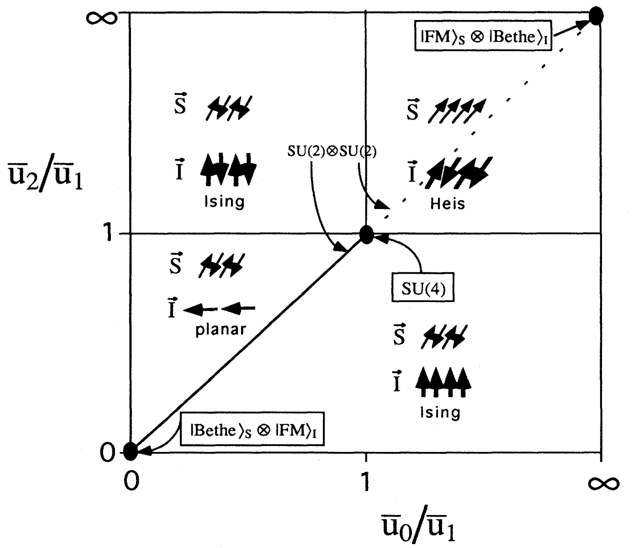

FIG. 1. Ground-state phase diagram for the Hamiltonian $\tilde{\mathcal{H}}$ of Eq. (22).

vector in a direction perpendicular to the chains, the $Q_a(\mathbf{i})$ are a set of local operators (e.g., magnetization) indexed by a, and $h_a^0(\mathbf{i})$ are a set of fields. As shown by Scalapino, Imry, and Pincus,[28] one can treat the interchain interactions by mean-field theory and thereby derive an expression for the full susceptibility $\chi_{ab}(\mathbf{q}_\perp,q_z)$ in terms of $\chi_{ab}^{1\mathrm{D}}(q_z)$, the susceptibility for the one-dimensional chains. The general result is

$$\chi(\mathbf{q}_\perp,q_z,\omega)=[\mathbb{1}-J_\perp(\mathbf{q}_\perp)\chi^{1\mathrm{D}}(q_z,\omega)]^{-1}\chi^{1\mathrm{D}}(q_z,\omega)\ , \tag{40}$$

where $J_\perp(\mathbf{q}_\perp)=\sum_{\boldsymbol{\delta}_\perp}J_\perp(\boldsymbol{\delta}_\perp)e^{-i\mathbf{q}_\perp\cdot\boldsymbol{\delta}_\perp}$ is the spatial Fourier transform of the interchain coupling matrix. [Note that the quantities χ, $J_\perp$, and $\chi^{1\mathrm{D}}$ in Eq. (40) are matrices.] This approximation also may be employed at finite temperature.

Consider now the $F\times A$ model discussed above. At finite temperature T, long-range ferromagnetic order is destroyed and the global SU(2)×SU(2) symmetry is restored. This means that the susceptibility matrix $\chi_{ab}^{1\mathrm{D}}(q_z,\omega)$ is diagonal, where a and b each represent any of the 15 operators $\{S^\alpha,I^\beta,S^\alpha I^\beta\}$.

As a crude approximation to the low-lying excitations of the $F\times A$ model, we assume independent ferromagnetic and antiferromagnetic spin waves as discussed above. In the ferromagnetic sector, we have $J_\parallel=4\ln(2)(t^\parallel)^2/\bar{u}_1$, owing to the reduction in the ferromagnon bandwidth due to the antiferromagnetic isospin correlations. The full dynamic susceptibility in the ferromagnetic sector is

$$\chi(q,\omega;T)=i\int_0^\infty dt\, e^{i\omega t}\langle[\mathcal{O}(q,t),\mathcal{O}^\dagger(-q,0)]\rangle\ , \tag{41}$$

with $\mathcal{O}=S^+$, and is well-approximated within the Schwinger-boson mean-field theory of Refs. 21 and 29 [a correction factor of $\frac{2}{3}$ has been included in Eq. (42), as discussed in Ref. 21]:

$$\chi_F''(q,\omega;T)=\frac{n_{(\omega-Qq^2)/2Qq}-n_{(\omega+Qq^2)/2Qq}}{12Q|q|}\ ,$$

$$n_k=\frac{1}{e^{\varepsilon_k/T}-1}\approx\frac{T}{\bar{\mu}+Qk^2}\rightarrow 2\pi S\delta(k)\ , \tag{42}$$

where $\varepsilon_k=\bar{\mu}+2Q(1-\cos k)\simeq\bar{\mu}+Qk^2$ is the mean-field Schwinger-boson dispersion, which at low temperatures ($J_\parallel\ll T$) is given by

$$Q=J_\parallel S\ ,\quad \bar{\mu}=T^2/4J_\parallel S^3 \tag{43}$$

(we have taken $k_B=\hbar=a=1$ here, where a is the lattice constant). The approximation in the second equation of Eq. (42) is valid for $T\gg J_\parallel$ and $Qk^2\ll T$. The static $k=0$ susceptibility is then

$$\chi_F'(0,0;T)=\frac{J_\parallel}{24T^2}+\cdots\ , \tag{44}$$

as was first derived by Takahashi in Ref. 30.

For the antiferromagnetic susceptibility, we appeal to the bosonization results of Schulz and of Eggert and Affleck,[31] who have computed the dynamic susceptibility of the $S=\frac{1}{2}$ quantum Heisenberg chain. In the vicinity of the antiferromagnetic ordering wave vector $k=\pi$ (of course, there is no true long-range order, even at $T=0$), one finds

$$\chi_A(\pi+q,\omega;T)=\frac{1}{\pi T}R\left[\frac{\omega+vq}{2\pi T}\right]R\left[\frac{\omega-vq}{2\pi T}\right]\ , \tag{45}$$

where

$$\begin{aligned}R(u)&=\int_0^\infty ds\frac{e^{isu}}{\sqrt{\sinh(s)}}\\&\simeq\sqrt{\pi/2|u|}\{1+i\,\mathrm{sgn}(u)\}\quad(|u|\gg1)\\&\simeq a_0+ia_1u+\cdots\quad(|u|\ll1)\ .\end{aligned} \tag{46}$$

Here $v=\pi J/2$ is the spin-wave velocity, $a_0\simeq4.44$, and $a_1\simeq6.16$. At long wavelengths and low frequencies ($v|q|,|\omega|\ll T$), one therefore has

$$\begin{aligned}\chi_A'(\pi+q,\omega;T)&\approx\frac{a_0^2}{\pi T}\ ,\\ \chi_A''(\pi+q,\omega;T)&\approx\frac{a_0a_1\omega}{\pi^2T^2}\ .\end{aligned} \tag{47}$$

Note that the static susceptibility diverges more weakly as $T\rightarrow0$ than in the ferromagnetic sector.

In the mixed F-A sector, where the operator in Eq. (41) is, e.g., $\mathcal{O}=S^xI^y$, we may use the assumed independence of ferromagnetic and antiferromagnetic magnons to write

$$\chi_{FA}(\pi+q,\omega)\approx\int d\mu\int d\nu\, S_F(q,\mu)S_A(\pi-q,\nu)\times\left\{\frac{1}{\omega+\mu+\nu+i\epsilon}-\frac{1}{\omega-\mu-\nu+i\epsilon}\right\}\ , \tag{48}$$

where $S(\omega)=\pi(1-e^{-\omega/T})\chi''(\omega)$. At low temperatures we find $\chi_{FA}'(\pi,0;T)\sim(J_\parallel T)^{-1/2}$, which diverges even more slowly than χ_A' in the $T\rightarrow0$ limit.

Thus, as the temperature is lowered, a transition to a ferromagnetic state should set in when $J_\perp\chi_F'=1$. The interchain interaction, given in Eq. (A5) in the Appendix, gives $J_\perp=J_\parallel[(t_1^\perp)^2+(t_2^\perp)^2]/4(t^\parallel)^2$, where $t_{1,2}^\perp$ are transverse hopping integrals defined in the Appendix. Thus we find $T_C\simeq\sqrt{J_\parallel J_\perp/24}$. The relation $T_C\propto\sqrt{J_\parallel J_\perp}$ was also found by Scalapino, Imry, and Pincus[28] *et al.* in their studies of anisotropic Heisenberg magnets. It is conceivable that at still lower temperatures a Néel ordering of the isospin variables occurs. From Eq. (A5), we expect that it is the I^x components which order antiferromagnetically, with coupling $3J_\perp$, leading to a Néel temperature $T_N\simeq3a_0^2J_\perp/\pi$.

VII. SUMMARY

We have introduced a new model of interacting electrons which describes quasi-one-dimensional hopping between doubly degenerate orbitals and local interactions parametrized by three pseudopotentials. At occupancy of one electron per site, we obtain a Mott insulator ground state with nearest-neighbor interactions of generalized spin variables. At special values of the pseudopotentials, the ground state of the model and certain elementary excitations are exactly known. These special points include the SU(4) Heisenberg antiferromagnet in the fundamental representation and the dual limits $\mathcal{H}_{F\times A}$ and $\mathcal{H}_{A\times F}$. We find a classical ground-state diagram which includes a large region of spin ferromagnetism and orbital antiferromagnetism. We believe this regime to be relevant for understanding the ferromagnetic properties of TDAE-C_{60}. The ferromagnetic behavior which arises in the limit $\bar{u}_1 \ll \bar{u}_0, \bar{u}_2$ is consistent with the physical picture of Seshadri *et al.*[7] A mean-field analysis of the interchain coupling in this regime predicts two transition temperatures: ferromagnetic spin ordering at $T_C \propto \sqrt{J_\parallel J_\perp}$ and orbital (isospin) antiferromagnetic ordering at $T_N \propto J_\perp$. This lower transition, to our knowledge, has not been observed experimentally. Perhaps the isospin ordering is superseded by an *isospin-Peierls* ordering (orbital dimerization) aided by the electron-phonon coupling. In that case, a signature for the isospin-Peierls effect should be present in x-ray scattering or in the phonon spectrum.

There are several open experimental issues which may be addressed by further extensions of our model. First is the role of orientational disorder. If the C_{60} molecules are frozen into a glassy orientational state,[32] this would introduce random couplings into the multicomponent superexchange model. Such perturbations could explain the weak ferromagnetism found in bulk magnetization measurements[3] versus the infinite (albeit reduced) frozen magnetic moment found in recent muon-spin-resonance (μ-SR) measurements.[4] In addition, Bloch's $T^{3/2}$ temperature dependence of the ordered moment found in Ref. 4, which holds up to $T \approx T_c$, is hard to reconcile with $J_\perp \ll T_c$ as expected in a quasi-one-dimensional system. Another open puzzle is the excessive entropy of the transition measured to be 34 kJ/mol (6 times larger than $R\ln 2 = 5.7$ kJ/mol, expected for localized spin-$\frac{1}{2}$ transitions.[33] We suggest that the measured entropy includes the disordering of both spins and isospins which can account for $2R\ln 2 = 11.4$ kJ/mol.

Note added in proof. Steve Kivelson (private communication) has proposed an interesting possibility that TDAE-C_{60} may have $\bar{u}_2 < \bar{u}_1$, $\bar{u}_2 < \bar{u}_0$. We can see in Fig. 1 that this implies a spin antiferromagnet and an *orbital* ferromagnet with a sizable magnetization along the c-axis direction. This possibility should be experimentally explored.

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APPENDIX: DETAILS OF THE SUPEREXCHANGE HAMILTONIAN

The matrices M^Λ appearing in Eqs. (13) and (18) can be written as direct products $M_{l\sigma,l'\sigma'} = Q_{ll'} \otimes R_{\sigma\sigma'}$, and are given by

$$\begin{aligned}
M^{(1,-)} &= \tfrac{1}{2} i\tau^y \otimes (\mathbb{1} - \tau^z) ,\\
M^{(1,0)} &= \frac{1}{\sqrt{2}} i\tau^y \otimes \tau^x ,\\
M^{(1,+)} &= \tfrac{1}{2} i\tau^y \otimes (\mathbb{1} + \tau^z) ,\\
M^{(2,-)} &= \tfrac{1}{2}(\mathbb{1} - \tau^z) \otimes i\tau^y ,\\
M^{(2,+)} &= \tfrac{1}{2}(\mathbb{1} + \tau^z) \otimes i\tau^y ,\\
M^{(0)} &= \frac{1}{\sqrt{2}} \tau^x \otimes i\tau^y .
\end{aligned} \tag{A1}$$

The superexchange Hamiltonian for the (ij) link may be written

$$\tilde{\mathcal{H}}(\mathrm{ij}) = \tilde{\mathcal{H}}^{(0)}(\mathrm{ij}) + \tilde{\mathcal{H}}^{(1)}(\mathrm{ij}) + \tilde{\mathcal{H}}^{(2)}(\mathrm{ij}) , \tag{A2}$$

where $\tilde{\mathcal{H}}^{(M)}(\mathrm{ij})$ is the superexchange Hamiltonian through the channel M ($M=0,1,2$) whose intermediate energy is $\bar{u}_M$. Assuming a hopping matrix for the bond (ij) is of the form in Eq. (16),

$$t_{l\sigma,l'\sigma'} = (t_1^\perp \mathbb{1} + t_2^\perp \tau^x)_{ll'} \delta_{\sigma\sigma'} , \tag{A3}$$

we find

$$\begin{aligned}
\tilde{\mathcal{H}}^{(0)} = -\frac{4}{\bar{u}_0} \{ & (t_1^\perp)^2 (I_\mathrm{i}^x I_\mathrm{j}^x + I_\mathrm{i}^y I_\mathrm{j}^y - I_\mathrm{i}^z I_\mathrm{j}^z + \tfrac{1}{4}) \\
& + t_1^\perp t_2^\perp (I_\mathrm{i}^x + I_\mathrm{j}^x) + (t_2^\perp)^2 (\tfrac{1}{4} + I_\mathrm{i}^x I_\mathrm{j}^x \\
& - I_\mathrm{i}^y I_\mathrm{j}^y - I_\mathrm{i}^z I_\mathrm{j}^z) \} \otimes (\tfrac{1}{4} - \mathbf{S}_\mathrm{i} \cdot \mathbf{S}_\mathrm{j})
\end{aligned} \tag{A4}$$

in the $M=0$ channel,

$$\begin{aligned}
\tilde{\mathcal{H}}^{(1)} = -\frac{4}{\bar{u}_1} \{ & (t_1^\perp)^2 (\tfrac{1}{4} - \mathbf{I}_\mathrm{i} \cdot \mathbf{I}_\mathrm{j}) + (t_2^\perp)^2 (\tfrac{1}{4} - I_\mathrm{i}^x I_\mathrm{j}^x \\
& + I_\mathrm{i}^y I_\mathrm{j}^y + I_\mathrm{i}^z I_\mathrm{j}^z) \} \otimes (\mathbf{S}_\mathrm{i} \cdot \mathbf{S}_\mathrm{j} + \tfrac{3}{4})
\end{aligned} \tag{A5}$$

in the $M=1$ channel, and

$$\begin{aligned}
\tilde{\mathcal{H}}^{2} = -\frac{2}{\bar{u}_2} \{ & (t_1^\perp)^2 (4 I_\mathrm{i}^z I_\mathrm{j}^z + 1) + 2 t_1^\perp t_2^\perp (I_\mathrm{i}^x + I_\mathrm{j}^x) \\
& - (t_2^\perp)^2 (4 I_\mathrm{i}^z I_\mathrm{j}^z - 1) \} \otimes (\tfrac{1}{4} - \mathbf{S}_\mathrm{i} \cdot \mathbf{S}_\mathrm{j})
\end{aligned} \tag{A6}$$

in the $M=2$ channel. Note that $\tilde{\mathcal{H}}(\mathrm{ij})$ becomes isotropic in both the spin and isospin channels when $t_2^\perp = 0$ and $\bar{u}_0 = \bar{u}_2$, when the intermediate two-electron states are organized into two threefold-degenerate multiplets, one a spin triplet and isospin singlet, the other a spin singlet and isospin triplet.

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