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Large-*J* approach to strongly coupled spin-orbital systems

Gia-Wei Chern and Natalia Perkins
Department of Physics, University of Wisconsin, Madison, Wisconsin 53706, USA

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We present an approach to study the ground-state and elementary excitations in compounds where spins and orbitals are entangled by on-site relativistic spin-orbit interaction. The appropriate degrees of freedom are localized states with an effective angular momentum J. We generalize J to arbitrary large values while maintaining the delicate spin-orbital entanglement. After projecting the intersite exchange interaction to the manifold of effective spins, a systematic $1/J$ expansion of the effective Hamiltonian is realized using the Holstein-Primakoff transformation. Applications to representative compounds Sr_2IrO_4 and particularly vanadium spinels AV_2O_4 are discussed.

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I. INTRODUCTION

Transition-metal compounds with partially filled orbitals have attracted considerable attention in recent years.[1] The interplay of spin and orbital degrees of freedom leads to a variety of interesting ground states and elementary excitations. Particularly, a long-range orbital order in the ground state makes possible the propagating orbital excitations, or orbitons,[2] in addition to the familiar spin waves. The intricate spin-orbital interaction dictated by the Kugel-Khomskii-type[3] superexchange (SE) Hamiltonian gives rise to a magnon spectrum which depends on the orbital order and vice versa. Meanwhile, interaction between the two types of quasiparticles leads to their scattering and decay.

Recently, there are a growing number of orbitally degenerate compounds whose elementary excitations cannot be described by the above paradigm based on magnons and orbitons. A common feature shared by these compounds is the presence of a strong relativistic spin-orbit (SO) interaction $V_{LS}=\lambda(\mathbf{L}\cdot\mathbf{S})$; most notable among them are vanadium and iridium oxides containing V^{3+} and Ir^{4+} ions, respectively.[4–7] To understand these systems, one should start with localized degrees of freedom which diagonalize the SO coupling.[7–9] Such atomic states are usually composed of complex orbitals and are characterized by an effective angular momentum J. The on-site spin-orbital entanglement leads not only to ground states with orbital-moment ordering,[8,10] but also to the appearance of composite quasiparticles which carry both spin and orbital characters.

Although several theoretical studies have been devoted to understanding of the ground-state properties in the limit of strong SO coupling,[8–12] very few works address the problem of elementary excitations. A straightforward approach is to use the magnon/orbiton description starting from the $\lambda=0$ ground state and to treat SO interaction as a perturbation.[13] However, as the large-λ ground state is fundamentally different from that of $\lambda=0$, such a formulation would not necessarily capture features like ordering of orbital moments. Another alternative is the magnetic-exciton model. Its application to vanadium spinels reveals an excitation spectrum characterized by transitions between states with different J_z.[14] Despite its success in providing a basic structure of excitations in the large λ limit, the magnetic-exciton model relies on an ill-controlled mean-field decoupling of the SE interaction. Besides, the model itself does not offer much insight to the ground-state structure.

In this Rapid Communication, we present a theoretical framework to study magnetically ordered ground state and its quasiparticles in systems with a dominating SO interaction. Instead of naively extending the length of the effective spin, our large-J generalization preserves the delicate entanglement between spins and orbitals. After obtaining the classical ground states in the $J\to\infty$ limit, a systematic $1/J$ expansion can be attained with the aid of Holstein-Primakoff transformation. Using the large-J approach, we examine two canonical examples: the iridium and vanadium compounds with effective spins $J=1/2$ and $J=2$, respectively. In the case of Ir^{4+} ion, the large-J approach is equivalent to conventional large-S method; the classical Hamiltonian has the same form as its quantum counterpart.

The situation for V^{3+} ion is more complicated. By minimizing the $J\to\infty$ limit of the effective Hamiltonian, we obtain a classical phase diagram of vanadium spinels. Our result provides a coherent explanation for the collinear and orthogonal antiferromagnetic orders observed in spinels ZnV_2O_4 and MnV_2O_4, respectively. The SE interaction in vanadium spinels has rather distinct characteristics in spin and orbital channels: while the spin exchange preserves a SU(2) symmetry, the orbital part resembles an anisotropic three-state Potts model. We find that the strong spatial dependence of the orbital exchange leads to a gapped excitation spectrum with a narrow bandwidth.

II. SPIN-ORBIT COUPLING AND EFFECTIVE SPINS

We start by describing the general large-J extension and then discuss specific applications to Ir^{4+} and V^{3+} ions with d^5 and d^2 configurations, respectively. In both cases, the orbital degrees of freedom of a partially filled t_{2g} triplet are described by an effective angular momentum of length $L'=1$. The true angular momentum is given by $\mathbf{L}=\alpha\mathbf{L}'$, where α of order one is the so-called covalency factor.[15] The SO interaction $V_{LS}=\alpha\lambda(\mathbf{L}'\cdot\mathbf{S})$ is diagonalized by the eigenstates of the "total angular momentum" $\mathbf{J}=\mathbf{L}'+\mathbf{S}$. Depending on the sign of α, the ground state has an effective angular momentum of length $J_{\text{eff}} \equiv L' \pm S$ and is separated from the excited states by a gap of order λ.

In order to carry out a controlled $1/J$ expansion of the effective Hamiltonian, we first generalize J to arbitrary values by considering a fictitious ion with N identical copies of t_{2g} triplets. The generalized SO interaction is $V_{LS} = \alpha\lambda \Sigma_{m=1}^{N} \mathbf{L}'_m \cdot \mathbf{S}_m$, where $\mathbf{L}'_m$ and $\mathbf{S}_m$ are orbital momentum and spin operators of the mth t_{2g} triplet, respectively. We further assume that only states which are symmetric with respect to the N degenerate t_{2g} triplets are allowed physical states. In the atomic ground state, each t_{2g} triplet has an angular momentum of length $J_m = J_{\text{eff}} = L' \pm S$, where $\mathbf{J}_m = \mathbf{L}'_m + \mathbf{S}_m$, and the symmetric sum $\mathbf{J} = \Sigma_{m=1}^{N} \mathbf{J}_m$ has the maximum length $J = NJ_{\text{eff}}$.

Single-ion operators such as orbital projections and spins are extended accordingly: $P_\alpha = \Sigma_{m=1}^{N} P_{m,\alpha}$ and $\mathbf{S} = \Sigma_{m=1}^{N} \mathbf{S}_m$. To obtain their representation in the $J = NJ_{\text{eff}}$ subspace, we expand its basis $|J, J_z\rangle$ with respect to the quantum number $J_{m,z}$ of the mth t_{2g} triplet

$$|J, J_z\rangle = \sum_{J_{m,z}} C_{J_z, J_{m,z}} |J_{\text{eff}}, J_{m,z}\rangle \otimes |J - J_{\text{eff}}, J_z - J_{m,z}\rangle, \tag{1}$$

where $C_{J_z, J_{m,z}}$ are the Clebsch-Gordon coefficients. By further expressing $|J_m, J_{m,z}\rangle$ in terms of $|L'_{m,z}, S_{m,z}\rangle$, one obtains the matrix elements of operators acting on the m-th t_{2g} orbitals. As the $J = NJ_{\text{eff}}$ subspace only contains permutationally symmetric states, the overall operator at a site is just N times that of a single t_{2g} triplet, e.g., $\mathbf{S} = N\mathbf{S}_m$.

Taking into account the intersite exchange interaction, the effective Hamiltonian of the angular momenta $\mathbf{J}_i$ in the $\lambda \to \infty$ limit is given by $H_{\text{eff}} \equiv \mathcal{P} H_{\text{SE}} \mathcal{P}$, where $\mathcal{P}$ is the projection operator onto the $J = NJ_{\text{eff}}$ manifold, and H_{SE} is the spin-orbital superexchange Hamiltonian.

Ir^{4+} *ions:* $L' = 1$ and $S = 1/2$. The iridium ions in compounds such as Sr_2IrO_4 are in the low spin $5d^5$ configuration with a positive covalency $\alpha \approx 1$.[15] The atomic ground state has an effective spin $J_{\text{eff}} = 1/2$. By introducing N identical t_{2g} replicas as described above, we generalize the effective spin to arbitrary large values $J = N/2$. Restricted to the lowest-energy manifold, we find $P_{xy} = P_{yz} = P_{zx} = 2J/3$, i.e., the electrons equally populate the three t_{2g} orbitals. The angular momentum and spin operators are given by

$$\mathbf{S} = -\mathbf{J}/3, \quad \mathbf{L}' = 4\mathbf{J}/3. \tag{2}$$

The magnetic moment of the Ir^{4+} ion is $\boldsymbol{\mu} = \mu_B(2\mathbf{S} + \alpha\mathbf{L}') \approx \frac{2\mu_B}{3}\mathbf{J}$. For compounds with 180° Ir-O-Ir bonds, the projected Hamiltonian

$$H_{\text{eff}} = \mathcal{J}_1 \sum_{\langle ij \rangle} \mathbf{J}_i \cdot \mathbf{J}_j + \mathcal{J}_2 \sum_{\langle ij \rangle} (\mathbf{J}_i \cdot \hat{\mathbf{r}}_{ij})(\mathbf{J}_j \cdot \hat{\mathbf{r}}_{ij}) \tag{3}$$

is dominated by a Heisenberg isotropic exchange plus a pseudodipolar interaction.[9] Here $\mathcal{J}_1 \approx 4\mathcal{J}$ and $\mathcal{J}_2 \approx 2\eta\mathcal{J}$ are effective exchange constants, $\eta = J_H/U$ is the ratio of Hund's coupling to on-site Coulomb repulsion U, and $\mathcal{J} = t^2/U$ defines the overall exchange energy scale. More importantly, we find that the large-J version of the effective Hamiltonian, and particularly the classical limit ($J \to \infty$), has the same form as the quantum Hamiltonian in Ref. 9. The large-J approach in this case thus is equivalent to the conventional large-S extension. In Sr_2IrO_4, the 'weak' ferromagnetic moment accompanying the ground-state antiferromagnetic order is explained by treating the spins as classical objects and taking into account the staggered rotations of the IrO_6 octahedra.[9] The large-J approach thus provides a theoretical basis for the classical treatment of the effective quantum Hamiltonian.

V^{3+} *ions:* $L' = 1$ and $S = 1$. Vanadium ions in cubic and spinel vanadates have a $3d^2$ configuration and a negative α. Consequently, the effective angular momentum minimizing the SO interaction has a length of $J_{\text{eff}} = 2$. The large-J generalization leads to the following orbital projection operator

$$P_{xy} = \frac{J(J-1) + J_z^2}{2(2J-1)}. \tag{4}$$

For projections $P_{yz/zx}$, we replace J_z by J_x and J_y, respectively. It could be easily checked that $P_{yz} + P_{zx} + P_{xy} = J$, which is just the total number of electrons $2N$. Similarly, projected to the $J = 2N$ subspace, the spin and orbital momentum operators are

$$\mathbf{S} = \mathbf{J}/2, \quad \mathbf{L}' = \mathbf{J}/2. \tag{5}$$

The angular-momentum quanta are evenly divided between the spin and orbital channels. The V^{3+} ion has a reduced magnetic moment $\boldsymbol{\mu} = \mu_B(1 - |\alpha|/2)\mathbf{J} \approx \frac{\mu_B}{2}\mathbf{J}$. Due to the spin-orbital entanglement of the $J = 2N$ states, the representation of the operator product $P_\alpha \mathbf{S}$ in this subspace is different from the matrix product of individual operators. Instead, we find

$$P_{xy}\mathbf{S} = \frac{(J-1)\mathbf{J}}{2(2J-1)} + \frac{J_z \mathbf{J} J_z}{2(J-1)(2J-1)}. \tag{6}$$

The cubic symmetry of the system allows us to obtain the expressions with P_{yz} and P_{zx} projections by simply replacing J_z by J_x and J_y, respectively.

The discussion so far has been general and is applicable to vanadium compounds with V^{3+} ions in an octahedral crystal field. We now focus on the case of spinels where the vanadium ions form the *pyrochlore* lattice, a network of corner-sharing tetrahedra. The 90° V-O-V bonds make direct exchange the primary mechanism of the SE interaction.[11,16] The resulting Potts-like orbital interaction leads to a highly anisotropic effective Hamiltonian in both coordinate and spin spaces.

Restricted to the $J = 2N$ manifold, the effective quantum Hamiltonian can be obtained by substituting the operator expressions (4)–(6) into the SE Hamiltonian for vanadium spinels. After introducing normalized classical spins $\hat{\mathbf{J}}_i \equiv \mathbf{J}_i/J$, the effective Hamiltonian in the classical limit $J \to \infty$ becomes

$$H_{\text{eff}} = \mathcal{J}_1 J^2 \sum_{\langle ij \rangle} (\hat{J}_{i,\gamma}\hat{J}_{j,\gamma})^2 + \mathcal{J}_2 J^2 \sum_{\langle ij \rangle} (1 + \hat{J}_{i,\gamma}^2)(1 + \hat{J}_{j,\gamma}^2)\hat{\mathbf{J}}_i \cdot \hat{\mathbf{J}}_j - \mathcal{J}_3 J^2 \sum_{\langle ij \rangle} [1 - (\hat{J}_{i,\gamma}\hat{J}_{j,\gamma})^2]\hat{\mathbf{J}}_i \cdot \hat{\mathbf{J}}_j. \tag{7}$$

Here the subscript $\gamma = \gamma(ij)$ denotes the x, y, or z component of the angular momentum $\mathbf{J}_i$ depending on whether the nearest-neighbor bond $\langle ij \rangle$ points along the $\langle 011 \rangle$, $\langle 101 \rangle$, or $\langle 110 \rangle$ directions, respectively. To lowest order in η, the effective exchange constants are $\mathcal{J}_1 \simeq \frac{1}{16}(1+5\eta)\mathcal{J}$, $\mathcal{J}_2 \simeq \frac{1}{16}(1-\eta)\mathcal{J}$, and $\mathcal{J}_3 \simeq \frac{\eta}{8}\mathcal{J}$.[11,16]

III. CLASSICAL GROUND STATE OF VANADIUM SPINELS

There has been much experimental effort to understand the properties of spinel vanadates AV_2O_4. For divalent ion A=Zn, Cd, and Mg, magnetic moments of V^{3+} ions are ordered collinearly in the ground state.[4,5] Theoretical models based on a large SO coupling have been proposed to explain the observed spin and orbital ordering.[8,11,12] However, a recent experimental characterization of another spinel MnV_2O_4 shows an orthogonal magnetic structure of vanadium spins.[6] The mechanisms that stabilize the orthogonal magnetic order remains unclear. Based on the large-*J* approach, here we propose a unifying model for the magnetic and orbital order in vanadium spinels.

The partially filled t_{2g} orbitals also couple to the distortions of the surrounding VO_6 octahedron. Indeed, both types of vanadium spinels undergo a cubic-to-tetragonal structural transition which accompanies the orbital ordering. The lattice distortion results in a splitting of the t_{2g} triplet: $V_{\text{JT}} = \Sigma_\beta \delta_\beta P_\beta$, where β runs over the three t_{2g} states. Using Eq. (4), the JT coupling can be recast into

$$V_{\text{JT}} = \delta_1 (J_x^2 + J_y^2 - 2J_z^2)/\sqrt{6} + \delta_2 (J_x^2 - J_y^2)/\sqrt{2}, \tag{8}$$

where (δ_1, δ_2) transforming as a doublet irreducible representation under the symmetry group O_h denote the tetragonal and orthorhombic distortions of the VO_6 octahedron, respectively.

Structural distortions compatible with the observed tetragonal symmetry (lattice constants $a=b>c$) in general involves $\mathbf{q}=0$ phonons with symmetries E_g and F_{1g}.[17] The E_g distortion gives rise to a uniform anisotropy δ_1, whereas softening of rotational F_{1g} phonons creates a staggered orthorhombic distortions $\pm\delta_2$ on VO_6 octahedra along the $[110]$ and $[1\bar{1}0]$ chains, respectively.[17] Classical ground state of a tetrahedron, the building block of the pyrochlore lattice, is obtained by minimizing the sum of effective Hamiltonian (7) and JT coupling (8). The resulting phase diagram is shown in Fig. 1(a).

The orthogonal magnetic order is stabilized at large values of the staggered F_{1g} distortion δ_2. Interestingly, the orthogonal structure also minimizes the $\mathcal{J}_1$ term of H_{eff}, which originates from the antiferro-orbital interaction in the original SE Hamiltonian. From Eq. (4), we see that d_{zx} and d_{yz} orbitals are fully occupied along the $[110]$ and $[1\bar{1}0]$ chains, respectively. This antiferro-orbital ordering is consistent with the low-temperature symmetry $I4_1/a$ of MnV_2O_4.[6] We note that the inclusion of SO interaction does not seem to affect the orbital-ordering pattern.[18] This could be attributed to the already mixed orbital states favored by a strong local trigonal distortion in MnV_2O_4.

On the other hand, the collinear antiferromagnetic order becomes the ground state when the lattice distortion is dominated by the E_g mode with $\delta_1 > 0$, which gives rise to an easy-axis anisotropy according to Eq. (8) The $\mathcal{J}_2$ term in Eq. (7) indicates that collinear spins parallel to the z-axis have the largest antiferromagnetic coupling on nearest-neighbor bonds lying in the xy planes. The ground state can then be viewed as a collection of antiferromagnetic Ising chains running along the $[110]$ and $[1\bar{1}0]$ directions [Fig. 1(c)], as was indeed observed in ZnV_2O_4.[4] The resulting orbital ordering has occupation numbers $n_{xy}=1$ and $n_{yz}=n_{zx}=1/2$, also consistent with the observed $I4_1/amd$ symmetry in ZnV_2O_4.[5] A similar staggered ordering of effective moment $\mathbf{J}$ is found in cubic vanadates at large λ.[10]

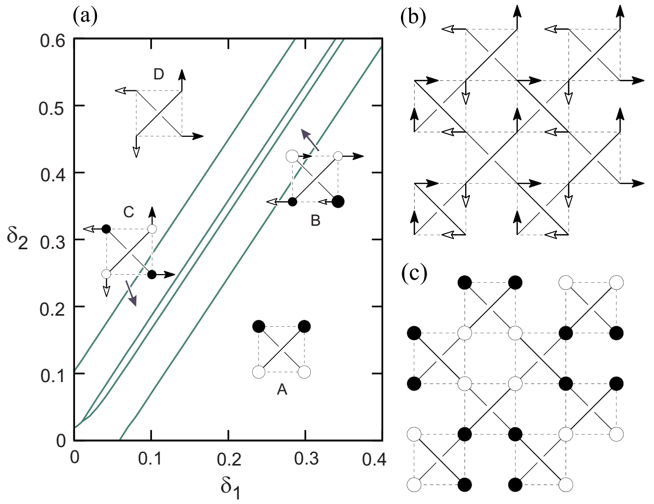

FIG. 1. (Color online) (a) Classical phase diagram of a tetrahedron. Black and white circles denote $\pm J_z$ components, respectively. δ_1 and δ_2 denote the uniform E_g and staggered F_{1g} distortions, respectively. A narrow low-symmetric phase with noncoplanar spins exists between phases B and C. (b) The $\mathbf{q}=0$ orthogonal order and (c) the $\mathbf{q}=(001)$ collinear order are observed in spinels MnV_2O_4 and ZnV_2O_4, respectively.

IV. HOLSTEIN-PRIMAKOFF TRANSFORMATION

Once the classical ground state is determined from the effective Hamiltonian, one can carry out a controlled $1/J$ expansion using the Holstein-Primakoff transformation,

$$J_z = J - a^\dagger a, \quad J_+ = \sqrt{2J - a^\dagger a}a, \quad J_- = J_+^\dagger. \tag{9}$$

The linear quasiparticle spectrum can then be obtained with the aid of Bogoliubov transformation; the calculated spectra for the orthogonal and collinear magnetic orders are shown by the solid lines in Fig. 2.

In the case of $\mathbf{q}=0$ orthogonal magnetic order, we obtain an acousticlike band whose gap scales as $\Delta \propto \sqrt{\delta_2 - \delta_{2c}}$ in the vicinity of $\delta_{2c} = 0.154\mathcal{J}$. The gapless mode at δ_{2c} signals the transition from phases D–C in Fig. 1(b). Although a similar gapped acoustic mode ($\Delta \approx 1.5$ meV) whose origin is attributed to the orthogonal order was indeed observed in MnV_2O_4,[6] the effect of the magnetic Mn^{2+} ions ($S=5/2$) remains to be clarified. On the contrary, all quasiparticle bands of the collinear magnetic order have an energy gap

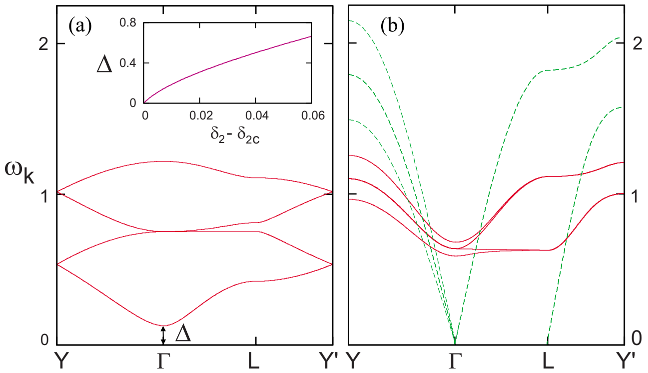

FIG. 2. (Color online) Quasiparticle spectra of vanadium spinels with (a) $\mathbf{q}=0$ orthogonal and (b) $\mathbf{q}=(001)$ collinear magnetic orders in the limit of strong spin-orbit coupling (solid lines). Also shown for comparison is the magnon spectrum of the collinear order without spin-orbit coupling (dashed lines). In obtaining these spectra, we have used $\delta_1=0$, $\delta_2=0.2\mathcal{J}$, and $\delta_1=\delta_2=0$ in (a) and (b), respectively. The symmetry points in k-space are $\Gamma=(0,0,0)$, $L=(\frac{1}{2},\frac{1}{2},\frac{1}{2})$, $Y=(0,1,0)$, and $Y'=(0,1,\frac{1}{2})$. The energy ω_k is measured in units of $\mathcal{J}\equiv t^2/U$. The inset in (a) shows the energy gap Δ as a function of staggered distortion δ_2.

$\Delta\approx 0.6\mathcal{J}$ even when $\delta_1=0$. This is because the staggered arrangement of frustrated bonds (parallel spins) in the $\mathbf{q}=(001)$ collinear order prevents the onset of the soft mode B shown in Fig. 1.

Also shown for comparison is magnon spectrum of the collinear order in the absence of SO coupling (dashed lines).[14] The Goldstone mode at the zone center reflects the global O(3) symmetry of the superexchange Hamiltonian in the $\lambda=0$ limit. The rather narrow bandwidth of the excitation spectrum in the large λ limit can be attributed to the fact that quasiparticles also carry orbital degrees of freedom whose Potts-like interactions are static.

V. CONCLUSION AND OUTLOOK

To summarize, we present an approach to the nontrivial problem of elementary excitations in systems with on-site spin-orbital entanglement. For such compounds, the ground-state magnetic order is usually accompanied by a long-range ordering of orbital moments. The bosonic elementary excitations resulting from deviations of the perfect order thus carry both spin and orbital degrees of freedom. Starting with the $J\rightarrow\infty$ limit, a systematic treatment of the elementary excitations can be realized through the standard $1/J$ expansion. The quasiparticle spectrum is obtained by diagonalizing the quadratic Hamiltonian. Higher-order effects such as quasiparticle scattering can also be studied using well developed methods from conventional large-S approach.

It is worth noting that spin-orbital entanglement could exist even in the absence of SO coupling due to the operator structure of SE Hamiltonian. For example, it is shown that composite spin-orbital fluctuations in cubic vanadates lead to orbital-disordered phase and violation of the Goodenough-Kanamori rules.[19] As the large-J Hamiltonian $H_{\text{eff}}=\mathcal{P}H_{\text{SE}}\mathcal{P}$ represents the lowest-order term in a $\mathcal{J}/\lambda$ expansion, another important front is to include the effects of finite λ. Higher-order terms can be obtained following the perturbation expansion of the SE Hamiltonian. In studying real compounds, λ is usually of the same order of exchange $\mathcal{J}$. Whether the excitations can be described by the large-J approach depends on the nature of the ground state. In the presence of orbital-moment ordering, we think the large-J method would be the appropriate choice.

Finally, we note that the classical effective Hamiltonian obtained in the $J\rightarrow\infty$ limit provides a practical working model for analyzing the experimental data. Furthermore, being a generic approach, our method can be easily adapted to other compounds where a large SO coupling dominates the low-energy physics.

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