Physical Review B (2009)-submetido Quantum disordered state in the frustrated quantum Heisenberg model on a stacked square lattice

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The ground-state phase diagram of the quantum spin-1/2 frustrated Heisenberg model in the presence of nearest-neighbor (J_1) and next-nearest-neighbor (J_2) interactions $(J_1 - J_2$ Heisenberg model) on a stacked square lattice, where we introduce an interlayer coupling through nearest-neighbor bonds of strength J_{\perp} , is studied within the framework of the differential operator technique. The Hamiltonian is solved by effective-field theory in cluster with two spins (EFT-2). We propose a functional for the free energy (similar to Landau expansion) to obtain the phase diagram in the (λ, α) space, where $\lambda = J_{\perp}/|J_1|$ and $\alpha = J_2/|J_1|$. Depending on the sign of J_1 , and values of λ and α , we obtain different collinear states, namely: a ferromagnetic $(J_1 < 0)$ collinear state (denoted by collinear ferromagnetic-**CF**) that is characterized by alternate up and down planes and an antiferromagnetic $(J_1 > 0)$ collinear (denoted by collinear antiferromagnetic-**CAF**) characterized by alternate up and down lines in all directions. For an intermediate region $\alpha_{1c}^{\mu}(\lambda) < \alpha < \alpha_{2c}^{\mu}(\lambda)$ $(\mu = F \text{ or } AF)$ we observe a quantum paramagnetic (QP) phase that disappears for λ above some critical value $\lambda_1 \simeq 0.32$ (0.54) when the nearest-neighbor interaction is ferromagnetic (antiferromagnetic). For $\alpha < \alpha_{1c}^{\mu}(\lambda)$ (and $\lambda < \lambda_1$) and $\alpha > \alpha_{2c}^{\mu}(\lambda)$ we have the F (AF) and CF (CAF) semi-classically ordered states, respectively. At $\alpha = \alpha^{\mu}_{1c}(\lambda)$ a second-order phase transition between the F (AF) and SL states occurs and at $\alpha = \alpha_{2c}^{\mu}(\lambda)$ a first-order transition between the F (AF) and CF (CAF) phases takes place. The boundaries between these ordered phases merge at the *critical end* point-**CEP** \equiv (λ_1, α_c), where $\alpha_c = 1/2$. Above this **CEP** there is again a direct first-order transition between the F (AF) and CF (CAF) phases, with a behavior described by the classical point $\alpha_c = 1/2$ independent of $\lambda \geq \lambda_1$ and sign of J_1 . In this work, we have predicted for first time a new quantum paramagnetic (or spin-liquid) state for the nearest-neighbor ferromagnetic interaction case.

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Low-dimensional spin systems are currently of strong interest because enhanced quantum fluctuations lead to unusual ground states and unusual low temperature properties. One of the models most studied in the past is that of a spin-1/2 Heisenberg antiferromagnet. The quantum Heisenberg antiferromagnet is a canonical model to describe quantum phase transitions driven by the interplay of competing interactions and quantum fluctuations. In particular, the two-dimensional (2D) quantum spin-1/2 Heisenberg model with competing nearest-neighbor (nn) and next-nearest-neighbor (nnn) antiferromagnet exchange interactions (i. e., *frustration*) on a square lattice (the $J_1 - J_2$ model) has been exhaustively studied by several methods¹⁻¹⁴, where the critical properties are relatively well known. In the absence of the nnn (next-nearest-neighbor) interactions (i.e., $J_2 = 0$), the system is not frustrated and the ground state possesses antiferromagnetic (AF) long-range order with wave vector $\mathbf{Q} = (\pi, \pi)$. The presence of the nnn interactions are expected to induce strong frustration to break the AF order, and that there is a quantum spin-liquid (SL) phase between α_{1c} and α_{2c} ($\alpha = J_2/|J_1|$). For $\alpha > \alpha_{2c}$ we have two degenerate collinear state which are the helical states with pitch vectors $\mathbf{Q} = (\pi, 0)$ and $(0, \pi)$ that are characterized by a parallel spin orientation of nearest neighbors in vertical (or horizontal) direction and an antiparallel spin orientation of nearest neighbors in horizontal (or vertical) direction and therefore exhibit Néel order within the initial sublattice A and B. The quantum disordered SL state is a singlet state with gapped excitations to the first triplet state¹⁴.

The critical properties of frustrated spin models strongly depend on the dimensionality (d), Hamiltonian symmetry (n) and spin (S). In particular, for the case of the 2d classical $(S \to \infty) J_1 - J_2$ Heisenberg (n = 3) model there is a consensus of the non existence of the SL state, with a first order transition at $\alpha_c = 1/2$ that separates the AF and collinear phases. Quantum fluctuations can modify drastically the ground state behavior, inducing,

for example, the SL state in the quantum spin-1/2 $J_1 - J_2$ Heisenberg model on a square lattice. For the one-dimensional (1d) case, this model with spin S = 1/2 does not have an AF ordered ground state, but exhibits a transition from a critical state to a dimer state at $\alpha_c^q = 0.241$ critical point¹⁶⁻¹⁸. The phase diagram in the $T - \alpha$ plane for the $J_1 - J_2$ Ising (n = 1) model on two and three-dimensional 3d lattices have been studied^{19,20}, where the SL state is not present. Although in three dimensions magnetic long-range order is more likely, a SL state may also be observed for frustrated 3d systems, e.g. for the Heisenberg antiferromagnet on the pyrochlore lattice²¹. The critical behavior of the square lattice version of the quantum spin-1/2 $J_1 - J_2$ Heisenberg model has been studied for many years, but very little has been done in the 3d case.

The quantum spin-1/2 Heisenberg model on the body-centered cubic (bcc) lattice has been studied recently²². It has been shown that the quantum $J_1 - J_2$ model on the bcc lattice does not have a quantum disordered SL phase, rather it exhibits a direct zero-temperature first order phase transition at $\alpha_c \simeq 0.7$ from the two-sublattice Néel phase to the so-called lamellar collinear state (sequences of up and down planes) driven by frustration J_2 . Later on, the 3d spin-1/2 $J_1 - J_2$ model on the simple cubic lattice has been studied by using effective field theory (EFT)²³ and a quantum first order transition was observed at $\alpha_c = 0.21$ that is smaller than the corresponding value of the classical $J_1 - J_2$ model $\alpha_c = 1/4$.

The case of ferromagnetic bonds in an antiferromagnetic matrix also have been discussed in connection with the proposal by Aharony and collaborators²⁴ to model localized holes in the CuO₂ planes by local ferromagnetic bonds between the copper spin to describe hightemperature superconductors. It was argued that random ferromagnetic bonds may influence the antiferromagnetic order drastically and may support the realization of a quantum spin liquid state^{25,26}. For small J_2 values, the 3d quantum spin-1/2 $J_1 - J_2$ antiferromagnetic has been used to describe the cuprates materials²⁷ and, materials Li₂VOSiO₄ and Li₂VOGeO₄ for the case of large J_2 (i.e., $J_2 \simeq J_1$). These two isostructural compounds are characterize by a layered structure containing V⁴⁺ (S = 1/2) ions²⁸. The structures of V⁴⁺ layer suggest that the superexchange is similar with a small interlayer coupling $J_{\perp} = \lambda J_1$, where $\lambda \simeq 10^{-2}$. In general, an interlayer coupling J_{\perp} may by relevant in real materials, it may have a crucial influence of the ground state magnetic ordering²⁹.

In this work, we consider the influence of such an interlayer coupling on the quantum

spin-1/2 $J_1 - J_2$ model on a simple cubic lattice, that is described by following Hamiltonian:

$$\mathcal{H} = \sum_{n} \left(J_1 \sum_{\langle i,j \rangle} \sigma_{in} \cdot \sigma_{jn} + J_2 \sum_{\langle \langle i,l \rangle \rangle} \sigma_{in} \cdot \sigma_{ln} \right) + J_\perp \sum_{i,n} \sigma_{in} \cdot \sigma_{in+1} \tag{1}$$

where $\sigma_{in} = (\sigma_{in}^x, \sigma_{in}^y, \sigma_{in}^z)$ are the spin-1/2 Pauli operators at site *i* in the *n*th-layer on the simple cubic lattice. The first and second sums run over the nearest-neighbor (nn) and next-nearest-neighbor (nnn) spin pairs, respectively, $J_1(J_2 = \alpha |J_1|)$ is the nn (nnn) coupling and $J_{\perp}(=\lambda |J_1|)$ the interlayer coupling. In the two-dimensional limit ($\lambda = 0$), in the ground state phase diagram the SL state is present, while in the isotropic 3d case ($\lambda = 1$) one may expect that this quantum disordered state is not observed.

The main motivation of this letter is to discuss the competition between the interlayer λ and frustration α parameters and to investigate their influence on the SL state. Here we consider the cases of ferromagnetic (F) and antiferromagnetic (AF) nn interactions that corresponds for $J_1(J_{\perp}) > 0$ and $J_1(J_{\perp}) < 0$, respectively.

The theoretical treatment of the frustrated quantum antiferromagnetism is far from being trivial. Many of the standard many-body methods, such as quantum Monte Carlo techniques, may fail or become computationally infeasible to implement if frustration is present due to the minus-sign problem. Hence, there is considerable interest in any method that can deal with frustrated spin systems. Recently²⁹, the model (1) has been studied for antiferromagnetic J_1 and J_{\perp} by using the coupled-cluster (CCM) and rotation-invariant Green's function (RGM) methods. It was found, that for a characteristic value $\lambda_1 \simeq 0.31(0.19)$ the quantum paramagnetic phase (i. e., the SL state) disappears using the RGM (CCM) approach. This considerable difference in that value for λ_1 further motivates us to study this issue by alternative methods. In this paper we will use the effective-field theory (EFT) in finite cluster to treat the model (1) and obtain the phase diagram at T = 0 (ground state). This method have been applied successfully to study a large variety of problems, in particular quantum models in arbitrary dimension³⁰⁻³² and it is able to study frustrated models^{30,31}.

The starting point for the EFT calculation is the choice of a finite cluster and obtain average of spin operators by using the Callen and Suzuki generalized relation (for more details, see Ref.³²). The EFT provides a hierarchy of approximations to obtain thermodynamic properties of magnetic models. On can continue this series of approximations to consider larger and larger clusters and as a consequence, better results are obtained. The exact solution would be obtained by considering an infinite cluster. However, by using relatively small clusters that contain the topology of the lattice, one can obtain a reasonable description of thermodynamic properties. The model (1) in two dimension ($\lambda = 0$) was recently treated by EFT in cluster with two spins (EFT-2)³⁰, where the phase diagram at T = 0 and finite temperature was obtained. In this limit of zero interlayer parameter λ we have the presence of the SL phase. For quantum spin systems, an appropriate choice for the ground state of the ordered phase is often a classical spin state. For example, in the case of the quantum AF order, we chose the classical Néel state for the ground state. While, the quantum collinear order we chose the classical states as shown in Fig. 1 dependent on the sign of J_1 .

In connection with the scenario of deconfined quantum criticality^{33,34} there has also been a considerable discussion of the nature of the quantum phase transition between the semiclassical Néel phase and the magnetically disordered (intermediate paramagnetic, i.e., the SL state) phase in the spin-1/2 $J_1 - J_2$ model with antiferromagnetic coupling $J_1 > 0^{9,13,14,35,36}$. On the other hand, the case of ferromagnetic coupling $J_1 < 0$ has been much less investigated so far. From the experimental side a new frustrated square lattice $J_1 - J_2$ system, Pb₂VO(PO₄)₂, was discovered, and thermodynamic measurements reveals the presence of ferromagnetic exchange³⁷. Quantum order from disorder occurs at low temperature, and the ground state observed below the Néel temperature $T_N \simeq 3.7K$ is a collinear antiferromagnet. Ferromagnetic nn exchange $J_1 \simeq -2K$ and antiferromagnetic nnn exchange $J_2 \simeq 6.5K$ was estimated, and, therefore, it corresponds to a new region of the $J_1 - J_2$ model phase diagram^{30,38-40} to be investigated.

The ground state of the classical frustrated Heisenberg model is more degenerate than the corresponding frustrated Ising model one. The average energy value at T = 0 are identical for the two classical frustrated models and is dependent on the α and λ parameters. Considering the collinear state, on the anisotropic simple cubic (sc) lattice, as made of alternate up and down **planes** (denoted of collinear ferromagnetic-CF state) with two degenerate wave vectors $\mathbf{Q} = \{(\pi, 0, 0), (0, \pi, 0)\}$, as shown in Fig. 1(a) for the vector state $\mathbf{Q} = (\pi, 0, 0)$, the corresponding average energy per spin is $E_o^{CF} \equiv \langle \mathcal{H} \rangle / N = 2\lambda J_1 - 4J_2$. In the For the collinear state characterized by alternate up and down **lines** in all directions (denoted by collinear antiferromagnetic-CAF state with two degenerate wave vectors $\mathbf{Q} = \{(0, \pi, \pi), (\pi, 0, \pi)\}$, as shown in Fig. 1(b) for the vector state $\mathbf{Q} = (\pi, 0, \pi)$, we have $E_o^{CAF} = -2\lambda J_1 - 4J_2$. The difference of the energy between this two ordered states $\Delta E_o \equiv E_o^{CF} - E_o^{CAF} = 4\lambda J_1$



FIG. 1: Ground state of the collinear ordered phases on a simple cubic lattice with (a) ferromagnetic nn interactions $J_1 < 0$ denoted by CF with vector $\mathbf{Q} = (\pi, 0, 0)$ and (b) antiferromagnetic nn interactions $J_1 > 0$ denoted by CAF with vector $\mathbf{Q} = (\pi, 0, \pi)$.

is dependent on the sign of J_1 . For the ferromagnetic case $(J_1 < 0)$ we have $\Delta E_o < 0$, and the configurations of spins for the ground state (global energy minimum) corresponds the CF state, while, the antiferromagnetic case $(J_1 > 0)$ we have $\Delta E_o > 0$, and the CAF phase corresponds the ground state. Therefore, the phase diagram for the 3d classical frustrated models is dependent on the sign of J_1 .

The average energy values at T = 0 for the F and AF states are given by $E_o^F = (4 + 2\lambda)J_1 + 4J_2$ and $E_o^{AF} = -(4 + 2\lambda)J_1 + 4J_2$, respectively. For these classical frustrated models the SL state between the F (AF) and collinear phases does not exist. The classical first-order transition point α_c that separates the F (AF) and CF (CAF) collinear phases is easily found by setting the energies between the ordered phases equal to each other. We obtain $\alpha_c \equiv (J_2/|J_1|) = 1/2$ that is independent of λ , i.e., it is the same value as for the classical $J_1 - J_2$ model on a square lattice ($\lambda = 0$). By using Monte Carlo simulation in the frustrated Ising model on a sc lattice, we have confirmed the stability of these collinear states (CF and CAF) dependent on the sign of J_1 . In the quantum model (1) corresponding semiclassical ground state phase should appear, however, with values for the order parameters less than the classical values due to quantum fluctuations. Hence we expect for the F case $(J_1 < 0)$ that there is the semi-classical collinear phase characterized by configuration of

spins corresponding to the CF state (Fig. 1(a)) and, for the AF case $(J_1 > 0)$ corresponding to the CAF state (Fig. 1(b)).

The quantum $J_1 - J_2$ Heisenberg model on the square lattice ($\lambda = 0$) has been extensively studied for the AF J_1 , see e.g. Refs.¹⁻¹⁴. In this case a SL state is found between α_{1c}^{AF} and α_{2c}^{AF} . The region of the SL phase $\Delta \alpha = \alpha_{2c}^{AF} - \alpha_{1c}^{AF}$ is decreasing with the increase of the spin value $S^{1,15}$, and most likely the SL phase is absent already for $S = 1^{15}$. Another important factor that decreases the region of the SL state is the sign of J_1 . Recently, the model (1) on a square lattice with F ($J_1 < 0$) and AF ($J_1 > 0$) nn interactions has been studied by various authors^{30,38-40}. In these papers the quantum phase transition points α_{1c}^F and α_{2c}^F between the ferromagnetic ground state and the SL phase and between the collinear phase and the SL phase, respectively, have been determined to $\alpha_{1c}^F = 0.4$ (Refs.^{38,39}), 0.43 (Ref.⁴⁰), 0.42 (Ref.³⁰), and to $\alpha_{2c}^F = 0.60 \sim 0.70$ (Refs.^{38,39}), 0.52 (Ref.⁴⁰), 0.56 (Ref.³⁰). Note that in Ref.³⁰ the corresponding values for the AF case have been determined to $\alpha_{1c}^{AF} = 0.28$ and $\alpha_{2c}^{AF} = 0.67$ using the same approximation as for the F case. Obviously, the region of the SL state $\Delta \alpha = 0.14$ for the F case is significantly smaller than for the AF case $\Delta \alpha = 0.39$. The shrinking of the SL region can be attributed to smaller quantum fluctuations in the F case in comparison to the AF case. The quantum phase transition from the F (AF) ordered state to the SL state at $\alpha = \alpha_{1c}^{\mu}$ (μ =F or AF) is of second order, while the transition from SL state to the CF (CAF) state at $\alpha = \alpha_{2c}^{\mu}$ is of first order. The increase of dimensionality, here represented in the model (1) by the interlayer parameter λ , is also an important factor that influences the quantum paramagnetic phase, and it will be analyzed in this work. In three dimensional lattice, we denote to the intermediate region between two ordered phases only of quantum paramagnetic (QP) phase.

For the EFT treatment of the model (1), we use the classical ground state as reference state. In order to illustrate the EFT, we choose a finite cluster with N = 2 spins that is schematized in Fig. 1 (a) for the collinear CF state. The Hamiltonian (1) for this cluster is written by

$$\mathcal{H}_2 = J_1 \sigma_a \cdot \sigma_b + C_a \sigma_a^z + C_b \sigma_b^z, \tag{2}$$

 $J_1\left(\sum_{i=16}^{18} \sigma_i^z + \lambda \sum_{i=14}^{15} \sigma_i^z\right) + J_2\left(\sigma_2^z + \sigma_3^z + \sigma_8^z + \sigma_9^z\right).$ As in our previous work^{23,30,31}, where the

effect of exchange anisotropy on the properties of the spin-1/2 $J_1 - J_2$ model on 2d and 3d lattices was studied, we again employ the EFT to investigate now the effect of spatial anisotropy, analytically we obtain an equation of state $m_{CF} = \Lambda_{CF}(m_{CF})$ for the CF phase (Fig. 1(a)) with the boundary condition: $m_p = m_{CF}$ for p = 1, 2, 4, 5, 6, 7, 8, 9, 18, 19, 20, 21, 22 and $m_p = -m_{CF}$ for p = 3, 10, 11, 12, 13, 14, 15, 16, 17, 23, 24, 25, 26.

Following the same procedure of Refs.^{23,30,31}, using the Hamiltonian (1) in cluster with two (N = 2) spins in the effective-field theory (EFT-2), for the collinear CAF state (Fig. 1(b)) we obtain $m_{CAF} = \langle \sigma_a^z \rangle = \Lambda_{CAF}(m_{CAF})$, with the boundary condition: $m_p = m_{CAF}$ for p = 3, 4, 5, 6, 7, 10, 11, 16, 17, 19, 20, 21, 22 and $m_p = -m_{CAF}$ for p =1, 2, 8, 9, 12, 13, 14, 15, 18, 23, 24, 25, 26. For the AF phase we have $m_{AF} = \Lambda_{AF}(m_{AF})$, with the boundary condition: $m_p = \left\langle \sigma_p^z \right\rangle = m_{AF}$ for p = 6, 7, 12, 13, 14, 15, 16, 17, 18, 23, 24, 25, 26and $m_p = \langle \sigma_p^z \rangle = -m_{AF}$ for p = 1, 2, 3, 4, 5, 8, 9, 10, 11, 19, 20, 21, 22. In the F case $(J_1 < 0)$, we have $m_p = m_F$ for all values of p = 1 - 26 and obtain an equation of state $m_F = \Lambda_F(m_F)$. Based on these equations of state, we may analyze the behavior of the respective order parameter in the AF (F) and collinear (CF and CAF) phases, for fixed values of the α and λ parameters, as a function of the temperature. The critical temperature $T_c(\alpha, \lambda)$ is determined by $m_{\mu} \to 0$ in the equation of state $m_{\mu} = \Lambda_{\mu}(m_{\mu})$. The first-order transition cannot be obtained on the basis only of the equations of state, since in this case one has $m_{\mu} \neq 0$ at the transition point. To solve this problem one needs to calculate the free energy for the F (AF), CF (CAF) and QP ($m_{\mu} = 0$) phases and to find a point of intersection. Following the same procedure from Ref.³⁰, after integration of the equations of state, we obtain a functional for the free energy $\Psi_{\mu}(m_{\mu})$.

The ground state (T = 0) phase diagram of the anisotropic 3d quantum spin-1/2 $J_1 - J_2$ Heisenberg model is shown in Fig. 2. It is dependent on the sign of J_1 . We observe five phases, namely: AF (antiferromagnetic), F (ferromagnetic), QP (quantum paramagnetic), CF and CAF phases. The F (AF) and QP phases are separated by a second-order transition line $\alpha_{1c}^{\mu}(\lambda)$, while the QP and collinear (CF and CAF) phases are separate by a first-order transition line $\alpha_{2c}^{\mu}(\lambda)$. The presence of the interlayer parameter λ has the general effect to suppress the QP phase. The QP region decreases gradually with the increase of the parameter λ , and it disappears completely at the *critical end point* $\mathbf{CEP} \equiv (\lambda_1, \alpha_1)$ where the boundaries between these phases merge. We find $\lambda_1 = 0.32$ (0.54) for the F (AF) case. Above this \mathbf{CEP} , i.e., for $\lambda > \lambda_1$, there is a direct first-order phase transition between the



FIG. 2: Ground state phase diagram in the (λ, α) plane for the quantum spin-1/2 $J_1 - J_2$ Heisenberg model on an anisotropic simple cubic lattice with (a) ferromagnetic (F) and (b) antiferromagnetic (AF) nn interactions. The first- and second-order transitions are indicated by the dashed and solid lines, respectively. The black point indicate the *critical end point* (CEP). We denote by QP, F, AF, CF and CAF the quantum paramagnetic, ferromagnetic, antiferromagnetic, collinear ferromagnetic and collinear antiferromagnetic phases, respectively.

AF and CAF as well as between the F and the CF phases, with a transition point $\alpha = 1/2$ independent of $\lambda \geq \lambda_1$ and also of the sign of J_1 . Such a direct first order transition was also observed for the classical $J_1 - J_2$ model and also for quantum $J_1 - J_2$ model on the body-centered cubic²² and the simple cubic²³ lattices.

The order parameter $m_{\mu}(T)$ ($\mu =$ F, AF) falls smoothly to zero when the temperature increases from zero to $T_c(\alpha, \lambda)$ when $\lambda < \lambda_1$ and $\alpha < \alpha_{1c}^{\mu}(\lambda)$ characterizing a second-order phase transition. On the other hand, for $\lambda < \lambda_1$ and $\alpha > \alpha_{2c}^{\mu}(\lambda)$ the magnetization curve $m_{\mu}(T)$ ($\mu =$ CF, CAF) may include an unstable solution in addition to the stable solution. Using Maxwell construction, that correspond then to that point in the phase diagram where the free energy between the QP and CF (CAF) phases are equal, we found the first-order transition temperature using the discontinuity of the magnetization at $T_c^*(\alpha, \lambda)$.

To summarize, has been confirmed in previous work^{12,30} that exchange anisotropy reduces the quantum fluctuations and leads to a shrinking of the QP region of the $J_1 - J_2$ model. In the present work, the quantum fluctuations in the $J_1 - J_2$ Heisenberg model are tuned by a nn interlayer coupling of strength J_{\perp} . Again the reduction of quantum fluctuations leads to a shrinking of the QP region. For the AF case we can compare our results obtained by effective-field theory with available results obtained by the CCM and RGM approaches²⁹. In Ref.²⁹ it was found that the QP phase disappears for $\lambda_1 \simeq 0.19$ (0.31) obtained by CCM (RGM). These results are in qualitative agreement with $\lambda_1 \simeq 0.54$ found in the present paper. However, the effective-field theory in cluster with two spins (EFT-2) seems to overestimate the value of λ_1 . On the other hand, the F case for $\lambda > 0$ has not been studied so far. It has been found in the present paper, see Fig. 2(a), that the QP phase found earlier for $\lambda = 0$ in Refs.^{30,38-40} may also exist if a finite interlayer coupling is present. In the case using cluster with N = 1 spin, denoted by EFT-1, this method can not be applied to treat the Heisenberg model. We have used also cluster with N = 4 spins (EFT-4) and the qualitative results are identical obtained in the present paper using EFT-2.

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