

Real-space perturbation theory for frustrated magnets: application to magnetization plateaus

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Abstract. We present a unified approach to the problem of degeneracy lifting in geometrically frustrated magnets with and without an external field. The method treats fluctuations around a classical spin configuration in terms of a real-space perturbation expansion. We calculate two lowest-order contributions for the Heisenberg spin Hamiltonian and use them to study the magnetization processes of spin- S triangular and kagomé antiferromagnets.

1. Introduction

A hallmark of geometrical magnetic frustration is degeneracy between lowest-energy spin configurations that is not related to any underlying symmetry. Generally, such degeneracy leads to enhanced fluctuations, thermal and quantum. Fluctuations play different roles depending on their strength. For spin models with large degeneracy and strong fluctuations, like spin-1/2 kagomé and pyrochlore antiferromagnets, magnetic ordering may be completely suppressed and a spin liquid state emerges at $T = 0$ [1]. On the other hand, if fluctuations are only moderate, they themselves provide an ordering mechanism commonly known as “order by disorder effect” [2, 3].

The analytic tool employed most of the time in theoretical studies of the order by disorder mechanism consists in the spin-wave expansion for a few reference states, see e. g. [3–12]. There is also an alternative route to finding the ground-state selection that can be loosely called the real-space perturbation theory (RSPT) [13–18]. Two main advantages of the latter approach are its simple analytic structure, at least in a few lowest orders, and description in terms of effective Hamiltonians operating in the manifold of classical ground states. This last feature allows to study the effect of fluctuations without any *ad-hoc* assumption about an outcome of the order from disorder selection. The same method can be also applied for investigation of the order by disorder effect produced by impurities and bond disorder [19, 20]. In Sec. 2 we present a general outline of the real-space perturbation approach for clean frustrated magnets, which has been so far missing in the literature. Then, we apply this theory in Sec. 3 to a prominent problem in frustrated magnetism—width of the 1/3 magnetization plateau in triangular and kagomé antiferromagnets in an external magnetic field. The rest of this section is devoted to a brief discussion of the classical ground-state constraints in geometrically frustrated magnets with and without an external magnetic field.



We consider the nearest-neighbor Heisenberg antiferromagnetic model in a magnetic field

$$\hat{\mathcal{H}} = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j - \mathbf{H} \cdot \sum_i \mathbf{S}_i. \quad (1)$$

The geometry of frustrated lattices allows decomposition of the nearest-neighbor spin Hamiltonian (1) into a sum over elementary frustrated units or blocks—triangles, tetrahedra etc. The minimum-energy condition corresponds, then, to a constraint on the total spin $\mathbf{L}_p = \sum_{i \in p} \mathbf{S}_i$ of every block. The constraint can be satisfied in multiple ways, which leads to degeneracy of classical spin configurations at $T = 0$.

Let us illustrate block decomposition of the spin Hamiltonian on the example of the triangular-lattice model. In this case, every exchange bond is shared between two triangular plaquettes, whereas every spin belongs to six triangles. By including proper compensation prefactors one can write a sum over spin triangles as

$$\hat{\mathcal{H}} = \sum_p \left[\frac{J}{4} \mathbf{L}_p^2 - \frac{\mathbf{H}}{6} \cdot \mathbf{L}_p - \frac{J}{4} (\mathbf{S}_{1,p}^2 + \mathbf{S}_{2,p}^2 + \mathbf{S}_{3,p}^2) \right], \quad \mathbf{L}_p = \mathbf{S}_{1,p} + \mathbf{S}_{2,p} + \mathbf{S}_{3,p}. \quad (2)$$

Minimization with respect to \mathbf{L}_p yields the ground-state constraint $\mathbf{L}_p = \mathbf{H}/(3J)$. The constraint fixes 3 out of 6 angles describing orientation of three sublattices. An additional continuous parameter is related to the breaking of the $SO(2)$ rotational symmetry in magnetic field. This leaves in total 2 free parameters, which describe degeneracy of the Heisenberg triangular antiferromagnet in an external field $0 < H < H_s = 9JS$ [21, 22]. For the real-space perturbation expansion we shall need a value of the local magnetic field acting on an individual spin in a ground-state spin configuration. It is obtained by relaxing for a moment the fixed spin length and differentiating the ground-state energy:

$$\mathbf{h}_i = -\frac{\partial E_{\text{g.s.}}}{\partial \mathbf{S}_i} = \frac{1}{2} JS \mathbf{n}_i \sum_{i \in p} 1 = 3JS \mathbf{n}_i, \quad \mathbf{n}_i = \mathbf{S}_i / |\mathbf{S}_i|. \quad (3)$$

Note, that only the last term in Eq. (2) contributes to \mathbf{h}_i because of the minimum condition with respect to \mathbf{L}_p . Thus, local fields in a classical ground-state configuration are always parallel to respective spins and their strength $H_{loc} = |\mathbf{h}_i| = 3JS$ does not depend on the site index i remaining constant for all fields $0 \leq H \leq H_s$. It is the site-independence of H_{loc} which allows to treat all classical ground states on equal footing within the RSPT.

The above approach with only minor modifications applies to many other geometrically frustrated models [23]. In particular, for Heisenberg antiferromagnets on kagomé and pyrochlore lattices, which consist of corner-sharing triangles and tetrahedra, the total spin of every block in the classical ground state is $\mathbf{L}_p = \mathbf{H}/2J$, whereas the local field amplitude is $H_{loc} = 2JS$. For the nearest-neighbor Heisenberg antiferromagnet on a face-centered cubic lattice, the spin Hamiltonian is represented as a sum over edge-sharing tetrahedra with the ground-state constraint $\mathbf{L}_p = \mathbf{H}/4J$ and the local field $H_{loc} = 4JS$.

2. Perturbation expansion around a classical ground state

Computation of the classical ground-state energy is equivalent to the mean-field approximation applied to a quantum spin Hamiltonian. Corrections to the mean-field approximation can be calculated by treating perturbatively correlations between spin fluctuations on adjacent sites. Construction of the perturbation expansion starts with rewriting the Hamiltonian in the local

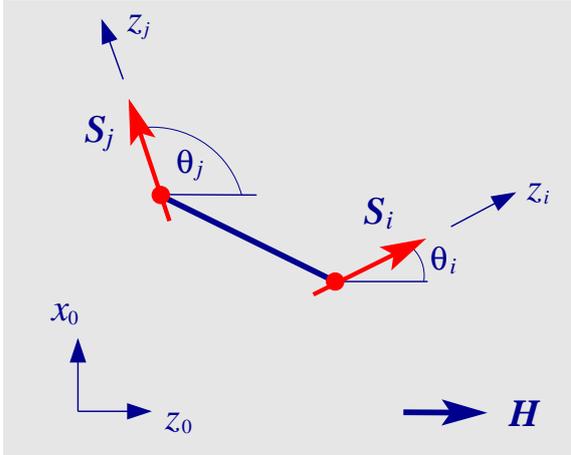


Figure 1. Choice of local axes and definition of angles for a non-collinear spin state in an external magnetic field.

spin frame, see Fig. 1, and collecting terms that depend on components of only one spin:

$$\begin{aligned} \hat{\mathcal{H}} = & E_{\text{class}} + H_{\text{loc}} \sum_i (S - S_i^z) + J \sum_{\langle ij \rangle} \left[S_i^y S_j^y + S_i^x S_j^x \cos \theta_{ij} + (S - S_j^z) (S - S_i^z) \cos \theta_{ij} \right. \\ & \left. + \sin \theta_{ij} (S_i^x S_j^z - S_i^z S_j^x) \right] + H \sum_i S_i^x \sin \theta_i. \end{aligned} \quad (4)$$

The z_i -axis on a given site is always pointing along \mathbf{S}_i , whereas orientation of x_i (y_i) is bond-dependent and assumed to lie in (be orthogonal to) the z_i - z_j plane with θ_{ij} being an angle between two spins on a given bond. All terms linear in S_i^x or S_i^y disappear due to the minimum energy condition. Then, dropping the classical energy constant E_{class} , we obtain

$$\begin{aligned} \hat{\mathcal{H}} = & H_{\text{loc}} \sum_i (S - S_i^z) + \hat{V}_1 + \hat{V}_2 + \hat{V}_3 + \hat{V}_4, \quad (5) \\ \hat{V}_1 = & -\frac{J}{4} \sum_{\langle ij \rangle} (1 - \cos \theta_{ij}) (S_i^+ S_j^+ + S_i^- S_j^-), \quad \hat{V}_2 = \frac{J}{4} \sum_{\langle ij \rangle} (1 + \cos \theta_{ij}) (S_i^+ S_j^- + S_i^- S_j^+), \\ \hat{V}_3 = & \frac{J}{2} \sum_{i,j} \sin \theta_{ij} (S_j^+ + S_j^-) (S - S_i^z), \quad \hat{V}_4 = J \sum_{\langle ij \rangle} (S - S_i^z) (S - S_i^z) \cos \theta_{ij}. \end{aligned}$$

The first term corresponding to the Zeeman energy in a local field H_{loc} is chosen as the unperturbed Hamiltonian $\hat{\mathcal{H}}_0$ with trivially calculated excited states, whereas bond terms \hat{V}_k are treated as perturbations. In principle, there is no an explicit small parameter for doing that. Still, since $h = O(zJ)$ and $\hat{V} = O(J)$, one can argue that such an approximation amounts to the $1/z$ expansion, with z being the coordination number.

In the following we focus on a quantum correction to the ground-state energy. For analogous RSPT treatment of thermal effects for classical models see Refs. [17, 20]. A noninteracting quantum ground state $|0\rangle$ coincides with a selected classical state and corresponds to a “fully saturated state” in the rotated basis: $S_i^+ |0\rangle = 0$. The perturbation \hat{V} can be treated using various forms of the perturbation expansion including the standard Rayleigh-Schrödinger theory. In this way we formulate a few simple rules that are used to identify nonzero contributions:

(i) Each term in the perturbation series is represented by a linked cluster ensuring the correct size scaling $\Delta E \propto N$. Every link corresponds to one of the perturbation terms \hat{V}_k acting on a specific lattice bond. The total number of links is equal to the order of RSPT expansion. Several links passing the same lattice bond are permitted.

(ii) The noninteracting ground state $|0\rangle$ is a vacuum for spin flips. Therefore, every term in the perturbation series starts and ends with creation $S_i^- S_k^-$ and annihilation $S_{i'}^+ S_{k'}^+$ of a pair of spin flips, corresponding to the action of the \hat{V}_1 operator.

(iii) All perturbations except of \hat{V}_3 conserve parity of the total number of spin-flips. Hence, every term in the ground-state energy expansion contains an even number of the \hat{V}_3 operators.

We now use the above rules for derivation of second- and third-order RSPT corrections. According to the rule (ii), the second-order correction has a very simple form and consists in the double action of the pair spin-flip operator \hat{V}_1 on the same bond:

$$|00\rangle \xrightarrow{S_i^- S_j^-} |11\rangle \xrightarrow{S_i^+ S_j^+} |00\rangle. \quad (6)$$

A graphical representation of this process is shown in Fig. 2(a). An intermediate state with two noninteracting spin flips has energy $2H_{loc}$. Calculating matrix elements of spin operators we find

$$E_2 = -\frac{J^2 S^2}{8H_{loc}} \sum_{\langle ij \rangle} (1 - \cos \theta_{ij})^2. \quad (7)$$

Apart from two unimportant terms that sum up to a state-independent constant, E_2 contains a biquadratic coupling $\sim \cos^2 \theta_{ij} = (\mathbf{n}_i \cdot \mathbf{n}_j)^2$ between nearest-neighbor spins. The energy (7) has a meaning of an effective Hamiltonian operating in the manifold of classical ground states parameterized by N unit vectors \mathbf{n}_i subject to the constraint. Therefore, a biquadratic term may arise even for $S = 1/2$ frustrated models. The negative sign in front of the biquadratic coupling favors the “most collinear” spin configurations among degenerate classical ground states. In many models with non-extensive ground state degeneracy, the order by disorder mechanism selects collinear or coplanar states [3–8]. This choice can be easily understood on the basis of the effective interaction (7), i.e. without doing any numerical computations that are required in the spin-wave theory.

An effective biquadratic interaction in the most general form (7) was first obtained by Heinilä and Oja [15] and, by now, has become a part of the verbal tradition in frustrated magnetism. A natural question to ask in this connection is whether it is appropriate to describe quantum effects in a frustrated magnet with the help of a biquadratic term perhaps with a phenomenological or fitted coefficient [24–26]. Feasibility of such a fit was questioned in Ref. [11], which showed that the spin-wave energy of the Heisenberg kagomé antiferromagnet in a magnetic field does not follow a simple cosine angular dependence expected from Eq. (7). A similar calculation for an anisotropic XY pyrochlore antiferromagnet has recently demonstrated that accuracy of the lowest order RSPT correction may improve significantly with increased anisotropy [20]. We shall now derive the complete third-order quantum correction, which has not so far been obtained in the literature and may help to further clarify the accuracy of Eq. (7).

For the frustrated lattices mentioned in Sec. 1, there are two types of third-order processes corresponding to dimer and triangle clusters that are shown in Fig. 2. The dimer processes, Fig. 2(b), are represented by the following diagram:

$$|00\rangle \xrightarrow{S_i^- S_j^-} |11\rangle \xrightarrow{S_i^z S_j^z} |11\rangle \xrightarrow{S_i^+ S_j^+} |00\rangle, \quad (8)$$

which describes subsequent action of \hat{V}_1 , \hat{V}_4 and again \hat{V}_1 . The energy correction from this process is

$$E'_3 = \frac{J^3 S^2}{16H_{loc}^2} \sum_{\langle ij \rangle} (1 - \cos \theta_{ij})^2 \cos \theta_{ij}. \quad (9)$$

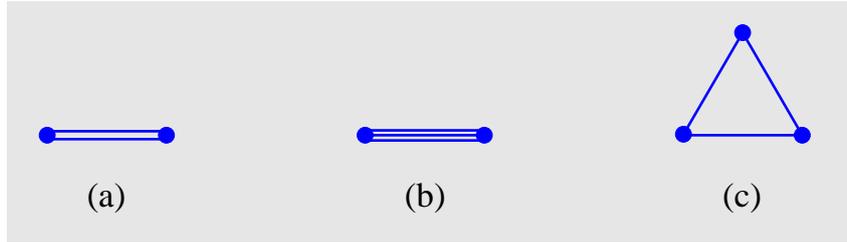


Figure 2. Linked clusters contributing in the second order (a) and in the third order, (b) and (c), of the real-space perturbation expansion.

The contribution E'_3 describes renormalization of the biquadratic exchange (7) by interaction between excited states.

For a triangular cluster (ijk), Fig. 2(c), a third-order process is described by the diagram

$$|000\rangle \xrightarrow{S_i^- S_j^-} |110\rangle \xrightarrow{S_j^+ S_k^-} |101\rangle \xrightarrow{S_k^+ S_i^+} |000\rangle \quad (10)$$

The corresponding energy shift is

$$E_\Delta = \frac{J^3 S^3}{32H_{loc}^2} (1 - \cos \theta_{ij})(1 + \cos \theta_{jk})(1 - \cos \theta_{ik}) e^{i(\varphi_{jk}^i + \varphi_{ik}^j + \varphi_{ji}^k)}, \quad (11)$$

where φ_{jk}^i is an angle between two planes formed by pairs of spins (i, j) and (i, k). Technically, the phase factors appear because of bond-dependent orientation of x_i and y_i axes in Eq. (4) and, as a result, in different phase factors of S_i^\pm operators in Eq. (5). Two reverse processes $ij \rightarrow jk \rightarrow ki$ and $ik \rightarrow kj \rightarrow ji$ have opposite phases and sum up into a real contribution. The phase factors disappear for locally coplanar spin structures such that spins on a triangular cluster form a single plane. Assuming further a coplanar configuration and summing over six possible processes for a given triangle, we finally obtain

$$E''_3 = \frac{J^3 S^3}{16H_{loc}^2} \sum_{\Delta} \left[(1 + \cos \theta_{ij})(1 - \cos \theta_{jk})(1 - \cos \theta_{ik}) + (1 - \cos \theta_{ij})(1 + \cos \theta_{jk}) \right. \\ \left. \times (1 - \cos \theta_{ik}) + (1 - \cos \theta_{ij})(1 - \cos \theta_{jk})(1 + \cos \theta_{ik}) \right]. \quad (12)$$

Real-space quantum corrections scale with specific powers of S . For example, $E_2, E''_3 \propto JS$ correspond to the harmonic spin-wave approximation, whereas $E'_3 \propto J$ is a nonlinear contribution. These terms constitute only a part of the respective spin-wave corrections, which naturally include graphs of arbitrary length. The number of relevant clusters quickly grows with the order of the real-space expansion and their evaluation beyond the fourth order may require numerical computations. Nonetheless, analytic calculations in higher orders can be used to identify the lowest-order perturbation processes that lift translational degeneracy for kagomé and pyrochlore antiferromagnets, see, for example, [18, 27–29].

3. Fractional magnetization plateaus in frustrated antiferromagnets

The general expressions for second- and third-order energy corrections derived in the previous section can be straightforwardly applied to the problem of order by disorder selection in magnetic field and, in particular, for calculation of the magnetization plateau width in triangular and kagomé antiferromagnets.

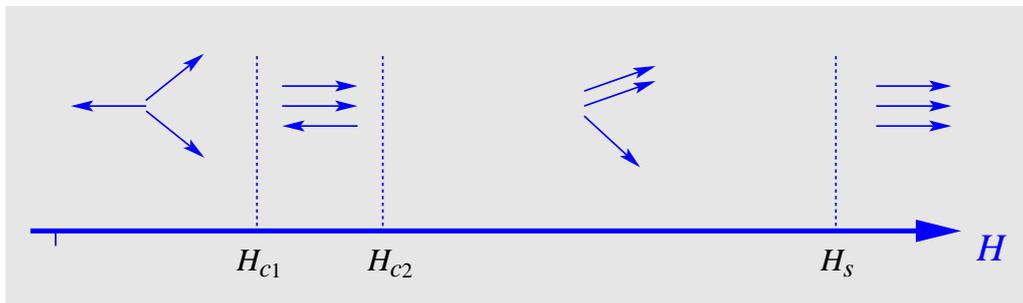


Figure 3. Three-sublattice ground-state spin configurations featured by triangular and kagomé antiferromagnets as a function of an external magnetic field: the Y-state for $H < H_{c1}$, the uud state for $H_{c1} \leq H \leq H_{c2}$ and the ∇ -state for $H > H_{c2}$.

3.1. Triangular Antiferromagnet

Degeneracy of the triangular-lattice antiferromagnet in an external field is determined by the classical constraint $\mathbf{S}_{\Delta} = \mathbf{H}/(3J)$ (Sec. 1), which can be satisfied in multiple ways by three magnetic sublattices. Among possible spin structures, thermal and quantum fluctuations select two coplanar states, the Y-state for $H < H_{c1}$ and the ∇ -state for $H > H_{c2}$, and a collinear uud state for $H_{c1} \leq H \leq H_{c2}$ [21, 22, 8], see Fig. 1. The collinear state is classically stable only for a single value of the external field $H_c = H_s/3 = 3JS$. Quantum fluctuations extend its presence to a finite range of fields around H_c and produce the $m = 1/3$ magnetization plateau. Calculation of the plateau width within the spin-wave theory is not entirely trivial and requires some sort of self-consistent approximation because of spurious negative-energy modes arising for the uud state at $H \neq H_c$, i.e. beyond the classical stability point [8, 30].

The problem of nonclassical ground state selection can be readily addressed by studying effective spin Hamiltonians obtained within the real-space perturbation approach. Using the expressions derived in Sec. 2, we have checked stability of the collinear spin structure with respect to small canting of three sublattices. Two critical fields, which bound the plateau region, are given by

$$H_{c1} = 3JS - \frac{J}{6S}, \quad H_{c2} = 3JS + \frac{J}{3} + \frac{J}{6S}. \quad (13)$$

Transitions at the plateau ends are continuous for all values of S . Note that H_{c1} and H_{c2} are shifted asymmetrically with respect to H_c , which differs qualitatively from a symmetric relation $H_c = (H_{c1} + H_{c2})/2$ obtained by using only a biquadratic term.

Let us now compare the above analytic expressions to available numerical results on the width of the $1/3$ -magnetization plateau in the triangular-lattice antiferromagnet. For $S = 1/2$, equation (13) gives $H_{c1} = 1.167$ and $H_{c2} = 2.167$ (in units of J), whereas the exact diagonalization study of finite clusters yields $H_{c1} = 1.381$ and $H_{c2} = 2.157$ [31]. For $S = 1$, our approximate analytic results are $H_{c1} = 2.833$ and $H_{c2} = 3.5$, which should be checked against numerical values $H_{c1} = 2.839$ and $H_{c2} = 3.552$ obtained in [32] by combination of the exact diagonalization and the coupled-cluster methods. Although such a remarkable agreement between numerics and a simple analytic theory is partly fortuitous, the above analysis shows the capability of the real-space perturbation theory to provide a quantitative description of quantum effects in frustrated magnets.

3.2. Kagomé Antiferromagnet

The kagomé antiferromagnet exhibits a much higher degeneracy of the classical ground states in comparison with the triangular-lattice model. The difference stems from loose connectivity

of triangle blocks in a kagomé structure, which form a network of corner-sharing triangles. As a result, infinitely many states with different translational patterns have the same classical energy. Still, at the level of each triangular block, short-range fluctuations select the same sequence of three-sublattice structures in a magnetic field as in the triangular antiferromagnet [11, 33]. Here we consider only this ‘short-range’ part of the quantum order by disorder effect in the kagomé antiferromagnet, because it is a much stronger effect than selection of a specific translation pattern and at the same time is responsible for the $m = 1/3$ magnetization plateau. Accordingly, we do not discuss nature of the plateau state: it can either possess some type of long-range order or remain in a spin liquid state.

Analysis of the combined contribution $E_2 + E'_3 + E''_3$ shows that the uud state is stable in a finite window of fields around $H_c = 2JS$:

$$2JS - \frac{J}{8} - \frac{J}{4S} \leq H \leq 2JS + \frac{3J}{8} + \frac{J}{4S}. \quad (14)$$

In the extreme quantum case of $S = 1/2$, the width of the magnetization plateau obtained from Eq. (14) is too large compared to available numerical results [34, 35]. On the other hand, for $S = 1$, our approximate calculation yields $H_{c1} = 1.625$ and $H_{c2} = 2.625$, which stand reasonably well against numerical DMRG values $H_{c1} \approx 1.67$ and $H_{c2} \approx 2.68$ [36]. Deficiency of the low-order RSPT calculation for the $S = 1/2$ kagomé antiferromagnet is not very surprising due to neglect of the tunneling processes between classical ground states that appear only in higher orders of the real-space expansion. The RSPT approach also fails to predict other fractional magnetization plateaus with $m = 5/9$ and $7/9$ determined by coherent hopping of spin flips around hexagons [34]. Still, a reasonable match between analytic and numerical values of the critical fields for $S = 1$ suggests a capability of the RSPT method to treat quantitatively even strongly frustrated spin models with $S > 1/2$.

4. Conclusions

We have given a general outline of the real-space perturbation theory for frustrated Heisenberg antiferromagnets. With little modifications the above approach also works for frustrated models with anisotropic exchange, see e.g. [20]. However, for the single-ion anisotropy, quantum fluctuations are already present at the level of the noninteracting Hamiltonian $\hat{\mathcal{H}}_0$ and this case requires a separate analysis. Let us also mention here that a somewhat different perturbation scheme was used by the authors of Ref. [18]. They have combined the local field term with the Ising part of the spin-flip interaction in Eq. (5) into a new noninteracting Hamiltonian $\hat{\mathcal{H}}_0 + \hat{V}_4$. This is equivalent to partial resummation of the perturbation series. Though, such resummation may be helpful for a certain class of problems, one disadvantage of this procedure is that the linked cluster representation of different perturbation terms is lost and one has to distinguish contractible and non-contractible clusters [18].

The RSPT expansion provides a simple qualitative description of the effect of quantum and thermal fluctuations on the ground state selection in geometrically frustrated magnets. The question of quantitative accuracy of the low-order RSPT results is, however, more delicate. Reasonable agreement between the third-order RSPT and numerical results for the plateau width in $S = 1$ triangular and kagomé antiferromagnets does not immediately imply that a next order correction would further improve the agreement. Nonetheless, the RSPT expansion carried out numerically to high orders by analogy with the standard Ising and dimer series expansion techniques for nonfrustrated magnets [37, 38, 39], should be able to address quantitatively many open questions and problems in the field of frustrated magnetism.

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