Semiclassical theory of the magnetization process of the triangular lattice Heisenberg model

Tommaso Coletta,[1] Tamás A. Tóth,[2] Karlo Penc,[3,4] and Frédéric Mila[5]

[1] *School of Engineering, University of Applied Sciences of Western Switzerland, CH-1950 Sion, Switzerland*
[2] *Haute école de gestion de Genève, University of Applied Sciences of Western Switzerland, CH-1227 Carouge, Switzerland*
[3] *Institute for Solid State Physics and Optics, Wigner Research Centre for Physics, Hungarian Academy of Sciences, H-1525 Budapest, P.O. Box 49, Hungary*
[4] *MTA-BME Lendület Magneto-optical Spectroscopy Research Group, 1111 Budapest, Hungary*
[5] *Institute of Physics, École Polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne, Switzerland*

(Received 20 May 2016; published 17 August 2016)

Motivated by the numerous examples of $1/3$ magnetization plateaux in the triangular-lattice Heisenberg antiferromagnet with spins ranging from $1/2$ to $5/2$, we revisit the semiclassical calculation of the magnetization curve of that model, with the aim of coming up with a simple method that allows one to calculate the full magnetization curve and not just the critical fields of the $1/3$ plateau. We show that it is actually possible to calculate the magnetization curve including the first quantum corrections and the appearance of the $1/3$ plateau entirely within linear spin-wave theory, with predictions for the critical fields that agree to order $1/S$ with those derived a long time ago on the basis of arguments that required going beyond linear spin-wave theory. This calculation relies on the central observation that there is a kink in the semiclassical energy at the field where the classical ground state is the collinear up-up-down structure and that this kink gives rise to a locally linear behavior of the energy with the field when all semiclassical ground states are compared to each other for all fields. The magnetization curves calculated in this way for spin $1/2$, 1, and $5/2$ are shown to be in good agreement with available experimental data.

DOI: 10.1103/PhysRevB.94.075136

I. INTRODUCTION

In strongly correlated electron systems quantum fluctuations are responsible for the manifestation of a variety of exotic behaviors. In the field of magnetic insulators, for instance, their effect can range from the stabilization of magnetic order to the emergence of nonmagnetic spin-liquid phases [1]. Of recent theoretical and experimental interest are the properties of frustrated magnetic insulators in external magnetic fields. In these systems, quantum fluctuations, which are enhanced by frustration, may lead to the presence of anomalies of the magnetization curve. Of specific relevance to our study are magnetization plateaux. These consist of a constant magnetization at a rational value of the saturation which persists over a finite field interval. While plateau states break the translational symmetry of the lattice, the nature of the plateau wave function greatly depends on the details of the model. Examples include crystals of purely quantum objects such as triplet excitations in ladder systems [2,3], crystals of more involved objects such as bound states of triplets as in the Shastry-Sutherland lattice [4], and valence-bond crystals as identified for the $S = 1/2$ Heisenberg antiferromagnet on the kagome lattice [5–7]. Such plateaux are usually referred to as "quantum" plateaux because the state which is stabilized has no classical analog.

By contrast, there are plateaux for which the magnetization pattern has a simple classical analog consisting of a crystal of down-pointing spins in a background of spins aligned with the magnetic field [8–12]. Such plateaux are sometimes referred to as "classical" plateaux. Given the essentially classical nature of such plateaux, it seems logical to expect that a purely semiclassical theory can be developed, and indeed, the first prediction of a $1/3$ plateau in the triangular lattice Heisenberg antiferromagnet by Chubukov and Golosov was based on semiclassical arguments [9]. They showed, going beyond linear spin-wave theory, that the $1/3$ plateau state with a three-sublattice up-up-down (uud) structure acquires a spin gap in a finite field range and that the critical fields at which the gap closes correspond to those at which the structure stops being collinear. Since the seminal work of Chubukov *et al.*, the existence of the $1/3$ plateau was confirmed numerically by exact diagonalizations of finite-size clusters for spin $S = 1/2$ [13] and $S = 1$ [14,15], as well as by the coupled-cluster expansion [16]. Moreover, several experimental realizations have been discovered: the compound Cs_2CuBr_4, although with an orthorhombic distortion [17–20], and the much closer realization of an ideal triangular lattice antiferromagnet $Ba_3CoSb_2O_9$ [21,22]. Both compounds are relevant for the spin $S = 1/2$ case. Additionally, we note that the compounds $Ba_3NiSb_2O_9$ and $RbFe(MoO_4)_2$ are other realizations of the same model, but this time the on-site magnetic moment is a spin $S = 1$ [14,15] and a spin $S = 5/2$ [23–27], respectively. In all of these systems magnetization measurements report the existence of a $1/3$ plateau.

Actually, Chubukov and Golosov did not calculate the magnetization curve outside the $1/3$ plateau using a semiclassical approach. Such a calculation was achieved years later in the case of the square-lattice antiferromagnet by Zhitomirsky and Nikuni [28], who showed that a semiclassical calculation of the magnetization curve is actually possible without going beyond the linear approximation if the magnetization is extracted from the derivative of the energy with respect to the field. The goal of the present paper is to show how this calculation can be extended to the case of the triangular lattice. This enterprise, which at first sight looks like a simple exercise, turned out to be far more subtle than expected and to raise a number of interesting questions. As we shall see, the magnetization curve calculated along the lines of Zhitomirsky and Nikuni is unphysical around the field where the classical ground state is the up-up-down state with magnetization $1/3$, and curing this unphysical behavior leads to an alternative semiclassical theory of the $1/3$ magnetization plateau entirely based on energy considerations which do not require going beyond linear order. The main conclusion is that it is indeed possible to calculate the magnetization curve of the triangular-lattice Heisenberg antiferromagnet (AFM) including the $1/3$ plateau within linear spin-wave theory. Remarkably enough, the critical fields derived by this alternative approach turn out to have the same value as those predicted by Chubukov and Golosov, whose approach required going beyond linear spin-wave theory.

To achieve this we will start by recalling the classical solution of the model (Sec. II) and the linear spin-wave prediction for the magnetization (Sec. III). Then we will discuss a phenomenological theory (Sec. IV) which we will then put on a more microscopic basis in the context of a variational argument (Sec. V). After comparing the results with available experiments (Sec. VI), we will conclude with a discussion of the validity and usefulness of the present results.

II. CLASSICAL SOLUTION

The Hamiltonian of the triangular-lattice Heisenberg antiferromagnet in a magnetic field is given by [29]

$$\mathcal{H} = \frac{J}{S^2}\sum_{\langle i,j\rangle}\mathbf{S}_i\cdot\mathbf{S}_j - \frac{H}{S}\sum_i S_i^z, \tag{2.1}$$

where the first sum is taken over all nearest neighbors of the triangular lattice [see Fig. 1(a)].

Up to a constant the Hamiltonian (2.1) can be rewritten as a sum over all triangular plaquettes of the lattice as

$$\mathcal{H} = \sum_p \frac{J}{4S^2}\left(\mathbf{S}_{p,1}+\mathbf{S}_{p,2}+\mathbf{S}_{p,3}-\frac{\mathbf{H}S}{3J}\right)^2, \tag{2.2}$$

with subscripts 1,2,3 denoting the three spins belonging to the plaquette p. At the classical level, when the spin operators are replaced by three-dimensional vectors of norm S, Eq. (2.2) indicates that the energy of the system is minimal when on all triangles of the lattice the total spin fulfills the constraint $(\mathbf{S}_{p,1}+\mathbf{S}_{p,2}+\mathbf{S}_{p,3}) = (S/3J)\mathbf{H}$. The resulting classical ground-state manifold is accidentally degenerate. For instance, both coplanar and umbrellalike configurations minimize the classical energy. Chubukov and Golosov showed that this accidental degeneracy is lifted at $T=0$ by quantum fluctuations in favor of the coplanar states [9]. The three-sublattice coplanar states stabilized in the linear spin-wave approximation can be parametrized by three angles measured with respect to the field direction [see Fig. 1(b)]. They are the Y state parametrized by $(\theta_1^Y,\theta_2^Y,\theta_3^Y)$ with $\theta_1^Y=\pi$ and $\theta_2^Y=-\theta_3^Y=\mathrm{acos}[(3J+H)/6J]$ for $0\leqslant H\leqslant 3J$ and the V state parametrized by $(\theta_1^V,\theta_2^V,\theta_3^V)$ with $\theta_1^V=-\mathrm{acos}[(-27J^2+H^2)/6HJ]$ and $\theta_2^V=\theta_3^V=\mathrm{acos}[(27J^2+H^2)/12HJ]$ for $3J\leqslant H\leqslant 9J$. When the field is at $1/3$ of the saturation value the Y and V states are identical to the uud structure with two spins pointing along the field and one pointing down on each triangular plaquette of the lattice.

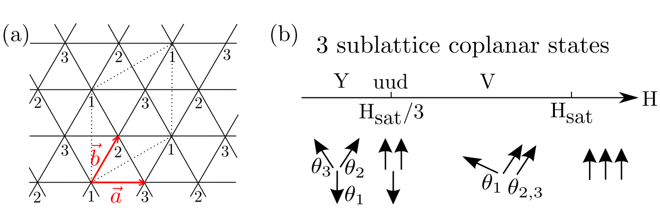

FIG. 1. (a) Triangular lattice and three-sublattice structure. The numbering indicates equivalent lattice sites. (b) Sketch of the three-sublattice Y and V coplanar structures at different field values.

In the next section we present some basic results of the spin-wave approximation for the Y and V coplanar structures.

III. LINEAR SPIN-WAVE APPROXIMATION

A. General formalism

The spin-wave approximation consists of the bosonic reformulation of the quantum spin problem in terms of Holstein-Primakoff (HP) particles which represent deviations from the underlying classical order and assuming these deviations to be small compared to the size of the classical moments. This approach is formalized in two steps: first, the quantum spin Hamiltonian is rewritten by expressing the spin operators in the local basis of the classical spin orientations denoted (x',y',z'). Supposing that the coplanar Y and V structures lie in the xz plane, this can be done as follows:

$$\begin{aligned} S^x_{\mathbf{R},i} &= \cos\theta_i S^{x'}_{\mathbf{R},i} + \sin\theta_i S^{z'}_{\mathbf{R},i},\\ S^y_{\mathbf{R},i} &= S^{y'}_{\mathbf{R},i},\\ S^z_{\mathbf{R},i} &= \cos\theta_i S^{z'}_{\mathbf{R},i} - \sin\theta_i S^{x'}_{\mathbf{R},i},\end{aligned} \tag{3.1}$$

where the angles θ_i parametrize the Y and V states, $\mathbf{R}$ is a vector of the superlattice, and $i=1,2,3$ denotes the sublattice [see Fig. 1(a)]. In this rotated frame, the classical ground state is ferromagnetic by construction.

Second, deviations from the classical order are expressed in terms of the Holstein-Primakoff [30] representation of spin operators. To next to leading order the expressions take the form

$$\begin{aligned} S^{x'}_{\mathbf{R},i} &= \frac{\sqrt{2S}}{2}(a_{\mathbf{R},i}+a^\dagger_{\mathbf{R},i}) - \frac{1}{4\sqrt{2S}}(n_{\mathbf{R},i}a_{\mathbf{R},i}+a^\dagger_{\mathbf{R},i}n_{\mathbf{R},i})+\cdots,\\ S^{y'}_{\mathbf{R},i} &= \frac{\sqrt{2S}}{2i}(a_{\mathbf{R},i}-a^\dagger_{\mathbf{R},i}) - \frac{1}{4i\sqrt{2S}}(n_{\mathbf{R},i}a_{\mathbf{R},i}-a^\dagger_{\mathbf{R},i}n_{\mathbf{R},i})+\cdots,\\ S^{z'}_{\mathbf{R},i} &= S - n_{\mathbf{R},i}.\end{aligned} \tag{3.2}$$

This transformation allows us to rewrite the quantum Hamiltonian (2.1) as a sum,

$$\mathcal{H} = \sum_{n=0}^{\infty}\mathcal{H}^{(n)}, \tag{3.3}$$

where $\mathcal{H}^{(n)} \propto S^{-n/2}$ contains only products of n bosonic operators. The first term of this series, $\mathcal{H}^{(0)}$, is the classical energy of the state around which fluctuations are considered. By construction, $\mathcal{H}^{(1)}$ vanishes identically since we expand around the three-sublattice coplanar spin configurations, which are minima of the classical energy. $\mathcal{H}^{(2)}$ describes the single-particle dynamics, and all higher-order terms in the expansion consist of many-particle interaction processes. Note that the bosonic representation is an exact mapping of the original quantum model. The spin-wave approximation consists of a truncation scheme based on an expansion in powers of $1/S$, the inverse of the magnetic moment being the small expansion parameter.

B. Ground-state energy in the harmonic approximation

At the harmonic approximation, which consists of truncating the expansion (3.3) to $n \leqslant 2$, the Fourier space expression of the fluctuation Hamiltonian is given by

$$\mathcal{H}^{(0)} + \mathcal{H}^{(1)} + \mathcal{H}^{(2)} = NE_{\rm cl} + \frac{1}{2S}\sum_{\mathbf{k}}[\mathbf{a}^\dagger_{\mathbf{k}} M_{\mathbf{k}}(H)\mathbf{a}_{\mathbf{k}} - \Delta_{\mathbf{k}}], \tag{3.4}$$

where $E_{\rm cl} = -3J/2 - H^2/18J$ is the classical energy per site of the three-sublattice coplanar states and N is the number of lattice sites. Since the states considered have three sites per unit cell, three distinct bosonic fields need to be introduced, and thus, the term $\mathbf{a}^\dagger_{\mathbf{k}}$ in Eq. (3.4) denotes the vector $(a^\dagger_{\mathbf{k},1}, a^\dagger_{\mathbf{k},2}, a^\dagger_{\mathbf{k},3}, a_{-\mathbf{k},1}, a_{-\mathbf{k},2}, a_{-\mathbf{k},3})$. $M_{\mathbf{k}}$ is a 6×6 matrix whose structure is detailed in Appendix A. The $1/S$ corrections to the classical energy are obtained by diagonalizing the fluctuation Hamiltonian (3.4) via a Bogolyubov transformation. The diagonal representation of (3.4) consists of a sum over three independent modes of free bosonic quasiparticles.

The ground-state energy per site corrected by fluctuations at $S = 1/2$ is depicted in Fig. 2. As can be seen, the energy presents a "kink" (discontinuity in the first derivative) at $H = H_{\rm sat}/3$, the value of the field at which the classical ground state is the uud state, as first noticed by Nikuni and Shiba [31]. This cusp, present for all values of the expansion parameter $1/S$, is most pronounced for $S = 1/2$. Quantum fluctuations are responsible for the emergence of the kink in the energy, whereas the classical energy is differentiable (see blue curve in Fig. 2).

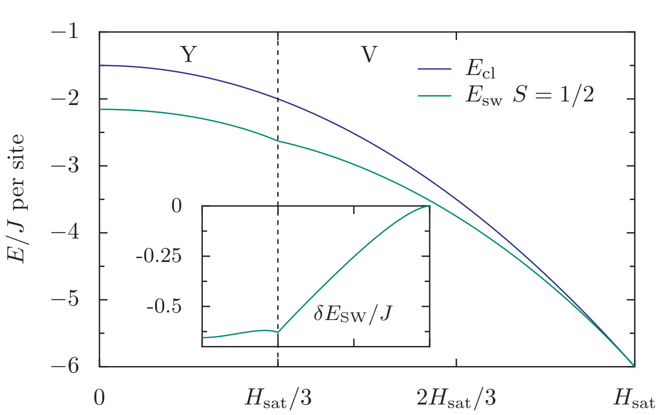

FIG. 2. Energy per site corrected by harmonic fluctuations for the coplanar Y- and V-type structures (green) and classical energy (blue). Inset: harmonic corrections to the classical energy. A kink is visible in the energy corrected by harmonic fluctuations at the field $H_{\rm sat}/3$.

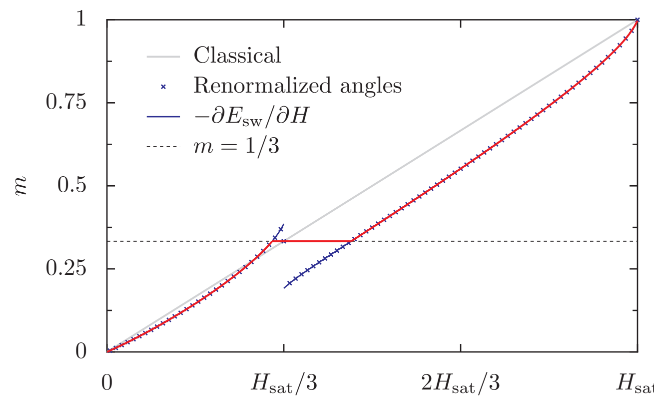

FIG. 3. Plots of the classical magnetization (gray solid line) and of the magnetization including corrections to first order in $1/S$ for $S = 1/2$ (blue curve). The $1/S$ corrections to the magnetization are computed in two equivalent ways: either as the derivative of the energy with respect to the magnetic field (blue curve) or by direct calculation taking into account the renormalization of the spin orientations (crosses). The overall $1/S$ magnetization curve obtained from our phenomenological approach is shown in red.

C. Magnetization curve

According to the Hellmann-Feynman theorem [32,33], the zero-temperature expression of the average magnetization per site is given by

$$m = -\frac{1}{N}\frac{\partial E_0}{\partial H}, \tag{3.5}$$

where N denotes the number of lattice sites and E_0 is the ground-state energy. To first order in $1/S$ the magnetization can be obtained from the derivative with respect to the field of the energy corrected by the zero-point motion [28] $E_0^{\rm harm}$ according to

$$m = -\frac{1}{N}\frac{\partial E_0^{\rm harm}}{\partial H}. \tag{3.6}$$

The average magnetization is presented in Fig. 3 for $S = 1/2$. When the $1/S$ corrections are included, the magnetization deviates from the straight-line classical behavior. As a consequence of the kink in the spin-wave energy as a function of the field, the magnetization displays a discontinuity at $H = H_{\rm sat}/3$. Associated with the discontinuity there is a "negative jump" in the magnetization occurring as the field is increased above $H_{\rm sat}/3$. This nonmonotonous behavior of the magnetization is, of course, unphysical and must be an artifact of the harmonic truncation of the $1/S$ expansion.

IV. PHENOMENOLOGICAL THEORY OF MAGNETIZATION

Since it is known from the work of Chubukov and Golosov that there is a plateau at 1/3, a phenomenological way to correct this unphysical aspect of the semiclassical magnetization of Fig. 3 consists of cutting the magnetization curve horizontally at the value $m = 1/3$. This phenomenological approach will be put on a more systematic basis in the next section. For the moment, let us prove that it leads to the same critical fields as those of Chubukov and Golosov.

In this phenomenological approach, the critical fields are defined by the intersection between the magnetization curve and the line $m = 1/3$. In order to extract the expressions for these critical fields one requires an analytic expression for the magnetization. An expression for the magnetization can be extracted from Eq. (3.6). This calculation, which turns out to be more technical in the case of states with multiple sites per unit cell for which the explicit expression of the Bogolyubov transformation is not known, is presented in Appendix A.

Alternatively, an analytic expression of the magnetization can be obtained by computing the quantum renormalization of the spin orientations following the procedure of Refs. [28,34]. In Appendix A it is shown that this method and the one presented in the previous paragraph yield rigorously the same results for the magnetization. For noncollinear states the angle renormalization procedure amounts to decoupling the cubic boson term $\mathcal{H}^{(3)}$ of the spin-wave expansion, which yields an effective linear boson contribution denoted $\mathcal{H}^{(3)}_{\rm eff}$. The cancellation of the overall linear boson term $\mathcal{H}^{(1)} + \mathcal{H}^{(3)}_{\rm eff}$ corresponds to a new stability condition which is fulfilled by a new set of renormalized angles. The renormalized spin orientations $\tilde{\theta}_i$ are expressed for each sublattice i as $\cos\tilde{\theta}_i = \cos\theta_i + c_i/S$, with the coefficients c_i given by

$$
\begin{aligned}
c_1^Y &= 0,\\
c_{2,3}^Y &= \cos\theta_2^Y(n_2 - m_{23} - \Delta_{23}) + \tfrac{1}{2}(-n_1 + m_{21} + \Delta_{21})
\end{aligned}
\tag{4.1}
$$

for the Y state and by

$$
\begin{aligned}
c_1^V &= -2\cos\theta_2^V(m_{21}+\Delta_{21}) + \cos\theta_1^V n_1 - \frac{3J}{H}(n_1 - 4n_2),\\
c_{2,3}^V &= -\frac{1}{2}\cos\theta_1^V(m_{21}+\Delta_{21}) + \cos\theta_2^V n_2 + \frac{3J}{2H}(n_1 - 4n_2)
\end{aligned}
\tag{4.2}
$$

for the V state, where in the above expressions we have introduced the following two-body averages computed in the harmonic ground state:

$$
n_i = \langle a^\dagger_{\mathbf{R},i} a_{\mathbf{R},i}\rangle, \quad m_{ij} = \langle a^\dagger_{\mathbf{R},i} a_{\mathbf{R}',j}\rangle, \quad \Delta_{ij} = \langle a_{\mathbf{R},i} a_{\mathbf{R}',j}\rangle,
\tag{4.3}
$$

with sites $(\mathbf{R},i)$ and $(\mathbf{R}',j)$ being nearest neighbors.

The expression of the magnetization per site in terms of the renormalized angles is

$$
m^{Y,V} = \frac{1}{3S}\sum_{i=1}^{3}\cos\tilde{\theta}_i^{Y,V}(S - n_i).
\tag{4.4}
$$

Collecting all terms up to order $1/S$ in (4.4) yields

$$
\begin{aligned}
m^Y &= \frac{H}{9J} + \frac{1}{3S}\left[-2\cos\theta_2^Y(m_{23} + \Delta_{23}) + m_{21} + \Delta_{21}\right],\\
m^V &= \frac{H}{9J}\left(1 - \frac{1}{S}(\Delta_{21} + m_{21})\right).
\end{aligned}
\tag{4.5}
$$

This expression of the magnetization is a function of the average quantities n_i, m_{ij}, and Δ_{ij}, whose field dependence is presented in Appendix A. The magnetization (4.5) is reported in Fig. 3 and coincides with that obtained from Eq. (3.6).

Now, Chubukov and Golosov [9] showed that, to leading order in $1/S$, the fields at which the Y and V structures become collinear [i.e., when the renormalized spin orientations, measured from the field direction, tend to $(\theta_1,\theta_2,\theta_3) = (\pi,0,0)$] correspond to the critical fields at which the gaps of the renormalized spectra of the uud state vanish (see Appendix A for more details). Below we show that the critical fields obtained by cutting the $1/S$ magnetization curve at the value $1/3$ are the same as those predicted by Chubukov and Golosov. For this purpose, let us introduce the quantities $H_{c1} = 3J + \alpha/S$ and $H_{c2} = 3J + \beta/S$ defined such that $m^Y(H_{c1}) = m^V(H_{c2}) = 1/3$. Evaluating the magnetization of the Y and V states at H_{c1} and H_{c2}, respectively, and expanding in powers of $1/S$ gives, to lowest order, [35]

$$
\begin{aligned}
m^Y(H_{c1}) &= \frac{1}{3} + \frac{\alpha}{9JS} + \frac{1}{3S}[-2(\bar{m}_{23} + \bar{\Delta}_{23}) + \bar{m}_{21} + \bar{\Delta}_{21}],\\
m^V(H_{c2}) &= \frac{1}{3} + \frac{\beta}{9JS} - \frac{1}{3S}(\bar{\Delta}_{21} + \bar{m}_{21}),
\end{aligned}
\tag{4.6}
$$

where the overbar denotes averages that are computed at $H = H_{\rm sat}/3$. Imposing $m^Y(H_{c1}) = m^V(H_{c2}) = 1/3$ and solving for α and β, we obtain

$$
\begin{aligned}
H_{c1} &= 3J\left(1 + \frac{2\bar{m}_{23} - \bar{\Delta}_{21}}{S}\right) = 3J\left(1 - \frac{0.084}{S}\right),\\
H_{c2} &= 3J\left(1 + \frac{\bar{\Delta}_{21}}{S}\right) = 3J\left(1 + \frac{0.215}{S}\right),
\end{aligned}
\tag{4.7}
$$

which correspond exactly to the same $1/S$ behaviors of the critical fields predicted by Chubukov and Golosov [9] (note that $\bar{m}_{21} = \bar{\Delta}_{23} = 0$; see Appendix A).

So we have shown that this very simple approach to determine the plateau boundaries, which consists of cutting the average magnetization to the value $1/3$, produces consistent results in the large-S limit. In the next section we present the formal justification for why the magnetization curve should be cut precisely at the value $m = 1/3$ as well as a novel perspective on the stabilization of the $1/3$ plateau which is based on the energetic comparison of the uud state with the other coplanar states.

V. VARIATIONAL THEORY OF MAGNETIZATION

To show that cutting the magnetization at $1/3$ is the right way to correct the unphysical behavior of the semiclassical magnetization of Fig. 3, let us first show that the existence of the kink in the energy curve corrected by harmonic fluctuations implies that the uud state will be stabilized over a finite field range. Our argument is the following: in the quantum Hamiltonian of the system (2.1) the total spin projection in the direction of the field is a conserved quantity. Hence the energies of the eigenstates of (2.1) depend linearly on the field. Now, the $1/S$ expansion of the Hamiltonian around the uud structure preserves this property even if the expansion is truncated at harmonic order. In the language of Holstein-Primakoff bosons this translates into the fact that $\sum_{\mathbf{R}}(-n_{\mathbf{R},1} + n_{\mathbf{R},2} + n_{\mathbf{R},3})$ commutes with the quadratic fluctuation Hamiltonian (where 1 denotes the sublattice site with spin down and 2 and 3 the sublattice sites with spin up). Therefore it is possible to determine the energy of the uud state, which can be computed to order $1/S$ only at $H_{sat}/3$, at other values of the field according to

$$E_{SW}^{uud}(H) = E_{SW}^{uud}(H_{sat}/3) - \frac{1}{3}\left(H - \frac{H_{sat}}{3}\right), \tag{5.1}$$

where $E_{SW}^{uud}(H_{sat}/3)$ is the energy per site of the uud state corrected by the zero-point fluctuations at $H = H_{sat}/3$ and $1/3$ is the average magnetization per site of the uud state.

The fact that the magnetization is strictly equal to $1/3$ in the uud state even when quantum fluctuations are included, as anticipated in Ref. [9], is not completely trivial since the local magnetizations are no longer equal to $\pm 1/2$ but are renormalized by quantum fluctuations. That this is true to order $1/S$ can be explicitly verified by calculating the local magnetizations at the harmonic order, which indeed satisfy $\langle -n_{\mathbf{R},1} + n_{\mathbf{R},2} + n_{\mathbf{R},3}\rangle = 0$. The proof that this is true to all orders is actually even simpler. Indeed, the full quantum Hamiltonian (2.1) can be split into the sum of two parts, $\mathcal{H}^z = J\sum_{\langle i,j\rangle} S_i^z S_j^z - H\sum_i S_i^z$ and $\mathcal{H}^{xy} = J\sum_{\langle i,j\rangle} S_i^x S_j^x + S_i^y S_j^y$. The uud state is an eigenstate of $\mathcal{H}^z$ with magnetization equal to $1/3$ of the saturation value and at the same time an eigenstate of $\sum_i S_i^z$ with eigenvalue $N/3$, while the term $\mathcal{H}^{xy}$ is to be viewed as a perturbation to $\mathcal{H}^z$. Since the commutator $[\mathcal{H}^{xy}, \sum_i S_i^z] = 0$ (i.e., the perturbation $\mathcal{H}^{xy}$ conserves the total spin projection in the z direction), any term generated in perturbation theory starting from the uud state has to be an eigenstate of $\sum_i S_i^z$ with the same eigenvalue $N/3$. So the resulting eigenstate of the full Hamiltonian still has a magnetization exactly equal to $1/3$ of the saturation value.

Now, since $E_{uud}(H_{sat}/3)$ is located at the position of the kink (and given the negative curvature of the energy as a function of the field; see Fig. 2), this construction indicates that in the vicinity of $H_{sat}/3$ the linear extrapolation of the uud-state energy (5.1) is lower than the energy of the neighboring Y and V states. Thus we predict that the energy as a function of the field has a linear behavior around the kink's location $H_{sat}/3$ and that the corresponding slope is $-1/3$ (see Fig. 4). This translates into a finite field interval of constant magnetization whose value is equal to $1/3$.

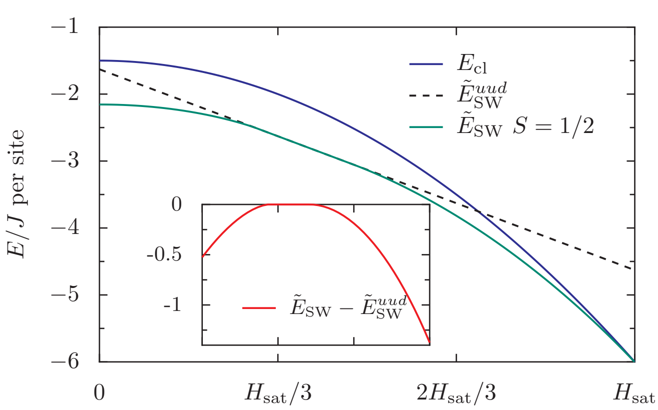

FIG. 4. The blue curve is the classical energy, and the green curve is the new energy curve constructed by extrapolating linearly the energy of the different structures. Inset: energy measured with respect to the energy of the uud state.

Simply using the linear extrapolation of the uud-state energy as a criterion for the stabilization of the plateau state overestimates the plateau width compared to Chubukov's result. The reason of this overestimation is that a similar extrapolation should also be used for the neighboring noncollinear Y and V states. Thus we propose to compare variationally the energy of all states as follows: let $|\phi_0\rangle$ denote the ground state (i.e., the Bogolyubov vacuum) of the harmonic fluctuation Hamiltonian around the state classically stable at $H = H_0$; then, the variational energy of this state (including harmonic fluctuations) at a different field is given by

$$E_0(H) = \langle\phi_0|\mathcal{H}(H_0)|\phi_0\rangle - (H - H_0)\langle\phi_0|\sum_i S_i^z/S|\phi_0\rangle. \tag{5.2}$$

A new energy curve $\tilde{E}(H)$ is obtained by comparing, at any given field H, the extrapolated energies of all structures. The resulting envelope is given by

$$\tilde{E}(H) = \operatorname*{Min}_{H_0}\left(\langle\phi_0|\mathcal{H}(H_0)|\phi_0\rangle - (H - H_0)\langle\phi_0|\sum_i S_i^z/S|\phi_0\rangle\right). \tag{5.3}$$

In this construction we allow a given coplanar state to be stabilized at a field which is different from the one for which it is the minimum of the classical energy. This mimics the mechanism by which quantum fluctuations renormalize the classical spin orientations. Given that both $\langle\phi_0|\mathcal{H}(H_0)|\phi_0\rangle$ and $\langle\phi_0|\sum_i S_i^z/S|\phi_0\rangle$ are quantities which are the sum of a classical contribution [of order $O(1)$] and of quantum corrections [of order $O(1/S)$], it can be shown that the value of H_0 minimizing Eq. (5.3) at a given H is such that the difference $H - H_0$ is also of order $1/S$ (see Appendix B for details). This can be understood simply by requiring that $\tilde{E}(H)$ must be equivalent to the classical energy in the limit $S \to \infty$, a condition that is fulfilled if the product $(H - H_0)\langle\phi_0|\sum_i S_i^z/S|\phi_0\rangle$ is a quantity which behaves as $1/S$. Therefore, to compare the energies of states to first order $1/S$, only the classical contribution to $\langle\phi_0|\sum_i S_i^z/S|\phi_0\rangle$ needs to be retained in Eq. (5.3).

In this construction the resulting energy curve $\tilde{E}(H)$ is strictly linear in the vicinity of $H_{sat}/3$. This behavior corresponds to the plateau stabilization (see Fig. 4). The plateau width obtained in this approach is reported as a function of $1/S$ in Fig. 5. The same plot also presents the plateau width estimates of Ref. [9] as well as the critical fields obtained numerically by cutting the magnetization curve at the value $1/3$. In all cases the agreement with Chubukov and Golosov's prediction is excellent for large S.

One should note that given the nontrivial field dependence of the magnetization curve corrected to first order in $1/S$, solving the equation $m(H) = 1/3$ for H yields solutions whose expression as a series in $1/S$ includes powers of $1/S$ greater than 1. This explains the discrepancy between the critical field prediction of this approach and that of Chubukov and Golosov for large values of $1/S$ (see Fig. 5). Nevertheless, Fig. 5 is the

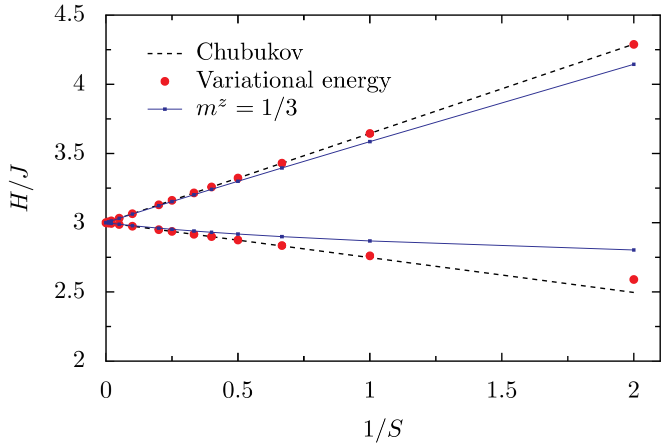

FIG. 5. Plot of the $1/3$ magnetization plateau width as a function of $1/S$ estimated by different approaches. Critical fields are determined by the condition $m = 1/3$ (blue curve) and the variational energy construction $\tilde{E}(H)$ (red points). The plot also presents the extension of the lowest-order Chubukov and Golosov prediction [9] to all S (dashed lines).

numerical confirmation that the leading $1/S$ behaviors are the same as predicted analytically.

VI. COMPARISON WITH EXPERIMENTS

To assess the validity of our magnetization curve construction, we compare it to recent magnetization measurements on different compounds which are the closest known experimental realizations of the Heisenberg model on the triangular lattice. Figure 6 compares the magnetization measurements for the compounds $Ba_3CoSb_2O_9$, $Ba_3NiSb_2O_9$, and $RbFe(MoO_4)_2$ (corresponding to a magnetic moment of $S = 1/2$, $S = 1$, and $S = 5/2$, respectively) to our $1/S$ prediction.

In spite of its simplicity, our theoretical prediction for the magnetization curve, which consists of cutting the $1/S$ magnetization at the value $m = 1/3$, yields results in good agreement with the experimental data for both the plateau width and position as well as for the magnetization curve away from the plateau. We stress, however, that our approach mainly provides an understanding of the plateau stabilization in the semiclassical approach. Recent numerical studies for spin $1/2$ [36,37] done in the context of the magnetization process of $Ba_3CoSb_2O_9$, including an XXZ anisotropy, are clearly more quantitative. For large spins, however, our semiclassical approach is expected to be accurate.

In that respect, we note that, in spite of the larger value of the magnetic moment, the agreement of our prediction with the measurements for $S = 5/2$ compound [Fig. 6(c)] is not as good as for the other compounds. We note, however, some discrepancies between the pulsed and static field measurements in $RbFe(MoO_4)_2$. Furthermore, for this compound, the saturation field is much smaller than that of the other systems. So, measured in units of the coupling constant, the effective temperature is much larger, and temperature effects cannot be neglected. The general trend that the plateau is a much smaller anomaly for larger spin is nevertheless supported by the experimental data.

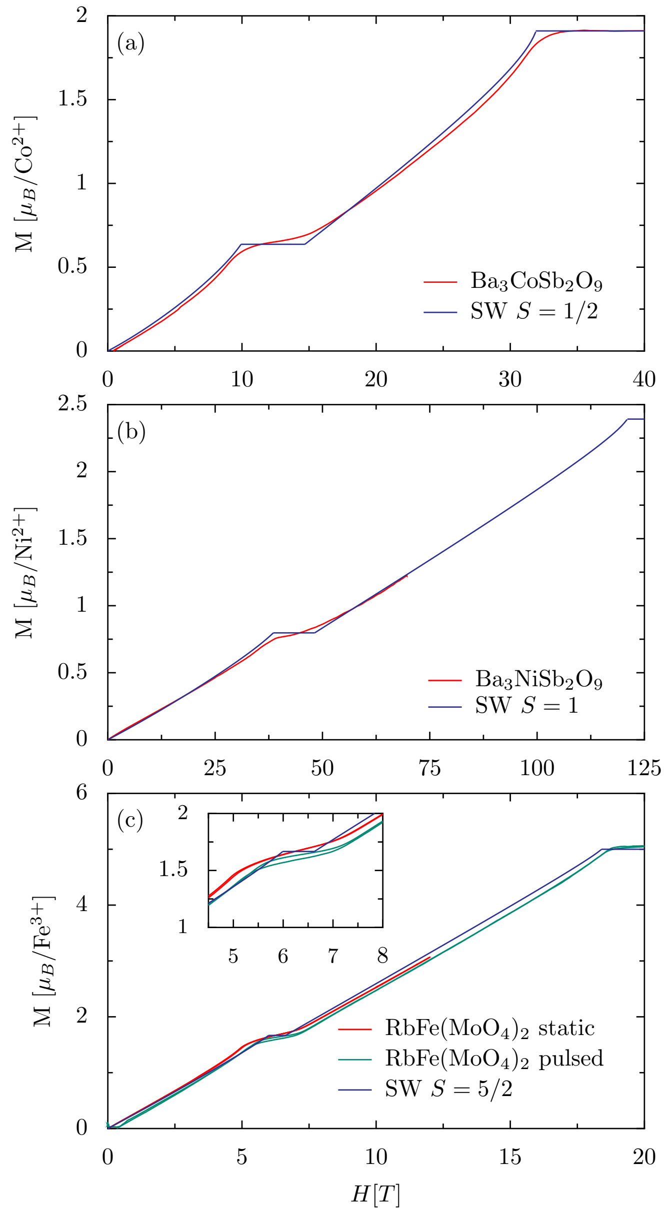

FIG. 6. Plot of the magnetization curve measurements for the compounds (a) $Ba_3CoSb_2O_9$ ($S = 1/2, T = 1.3$ K, powder sample) [21], (b) $Ba_3NiSb_2O_9$ ($S = 1/2, T = 1.3$ K, powder sample) [14] (b), and (c) $RbFe(MoO_4)_2$ ($S = 5/2, T = 1.3$ K, pulsed field) [25] and $RbFe(MoO_4)_2$ ($S = 5/2, T = 1.55$ K, static field) [23] and of the $1/S$ prediction at different values of S.

VII. CONCLUSION

In conclusion, we have shown that a semiclassical calculation of the magnetization curve of the Heisenberg model on the triangular lattice which includes the plateau at $1/3$ and which is correct to order $1/S$ can be simply obtained in two steps: (i) calculate the magnetization as minus the derivative of the harmonic energy with respect to the field and (ii) cut this curve by a horizontal line at $1/3$. The justification of cutting this curve at $1/3$ relies in an essential way on the presence of a kink in the semiclassical energy for the field at which the uud state is stabilized. Thus this simple method can be generalized to other models, with step (ii) being replaced by a cut around each point where the semiclassical energy has a kink, with the corresponding magnetization.

Of course, this simple approach does not give access to all details of the magnetization curve. In particular, it leads to cusps with finite slopes at the plateau boundaries, whereas general arguments suggest that the transition into the plateau state should be either of the first order accompanied by a magnetization jump or continuous and display a logarithmic singularity with an infinite slope since it belongs to the same universality class as the transition into the saturated phase [28,38–40]. To access these details requires to go beyond the linear order in the spin wave expansion.

However, as demonstrated by the comparison with experimental data, the present theory is quite accurate even for $S = 1/2$, and it would presumably take experiments at very low temperature in highly isotropic systems to actually observe significant deviations from the present theory, provided, of course, the system does not realize nonclassical ground states on the way to polarization. Considering the difficulty in pushing spin-wave theory beyond linear order, it is our hope that the present approach, which relies on only the elementary linear spin-wave theory, will be useful to both experimentalists and theorists in the investigation of the magnetization process of frustrated quantum magnets.

ACKNOWLEDGMENTS

We acknowledge many valuable discussions with S. Korshunov at an early stage of this project. We are indebted to the authors of Refs. [14,21,25] for providing the magnetization measurement data presented in Fig. 6. This work has been supported by the Swiss National Science Foundation and by the Hungarian OTKA Grant No. K106047.

APPENDIX A: SPIN-WAVE THEORY

This section presents the explicit expression of some results of the linear spin-wave approximation for a generic three-sublattice coplanar state, as well as some aspects of the calculation to higher order referred to in the text.

1. Linear spin-wave approximation and $1/S$ magnetization

The block structure of the harmonic fluctuation matrix $M_{\mathbf{k}}$ entering Eq. (3.4) is

$$M_{\mathbf{k}} = \begin{pmatrix} \bar{\bar{A}}_{\mathbf{k}} & \bar{\bar{B}}_{\mathbf{k}} \\ \bar{\bar{B}}_{\mathbf{k}} & \bar{\bar{A}}_{\mathbf{k}} \end{pmatrix}, \tag{A1}$$

with

$$\bar{\bar{A}}_{\mathbf{k}} = \begin{pmatrix} A & \gamma_{\mathbf{k}}^{\star} D & \gamma_{\mathbf{k}} H \\ \gamma_{\mathbf{k}} D & B & \gamma_{\mathbf{k}}^{\star} F \\ \gamma_{\mathbf{k}}^{\star} H & \gamma_{\mathbf{k}} F & C \end{pmatrix},$$
$$\bar{\bar{B}}_{\mathbf{k}} = \begin{pmatrix} 0 & \gamma_{\mathbf{k}}^{\star} E & \gamma_{\mathbf{k}} I \\ \gamma_{\mathbf{k}} E & 0 & \gamma_{\mathbf{k}}^{\star} G \\ \gamma_{\mathbf{k}}^{\star} I & \gamma_{\mathbf{k}} G & 0 \end{pmatrix}. \tag{A2}$$

The coefficients entering Eq. (A2) are

$$\begin{aligned} A &= [-3J(\cos\theta_{1,2} + \cos\theta_{1,3}) + H\cos\theta_1], \\ B &= [-3J(\cos\theta_{1,2} + \cos\theta_{2,3}) + H\cos\theta_2], \\ C &= [-3J(\cos\theta_{1,3} + \cos\theta_{2,3}) + H\cos\theta_3], \\ D &= J(\cos\theta_{1,2} + 1)/2, \quad E = J(\cos\theta_{1,2} - 1)/2, \\ F &= J(\cos\theta_{2,3} + 1)/2, \quad G = J(\cos\theta_{2,3} - 1)/2, \\ H &= J(\cos\theta_{1,3} + 1)/2, \quad I = J(\cos\theta_{1,3} - 1)/2. \end{aligned} \tag{A3}$$

where $\theta_{i,j} = \theta_i - \theta_j$ is the difference between the spin orientations on sublattices i and j (see Fig. 1). The geometrical coefficient $\gamma_{\mathbf{k}}$ is given by

$$\gamma_{\mathbf{k}} = (e^{i\mathbf{ka}} + e^{-i\mathbf{kb}} + e^{i\mathbf{k}(-\mathbf{a}+\mathbf{b})}) \tag{A4}$$

for the triangular-lattice basis vectors **a** and **b** defined in Fig. 1(a). The additional term $\Delta_{\mathbf{k}}$ in Eq. (3.4) is equal to the trace of $\bar{\bar{A}}_{\mathbf{k}}$, $\Delta_{\mathbf{k}} = \mathrm{Tr}[\bar{\bar{A}}_{\mathbf{k}}]$.

The Bogolyubov transformation which diagonalizes (3.4) consists of a 6×6 momentum dependent matrix $T_{\mathbf{k}}$ with block structure

$$T_{\mathbf{k}} = \begin{pmatrix} U_{\mathbf{k}} & V_{\mathbf{k}} \\ V_{\mathbf{k}} & U_{\mathbf{k}} \end{pmatrix}. \tag{A5}$$

For any value of momentum, $T_{\mathbf{k}}$ simultaneously fulfills the conditions that (i) $T_{\mathbf{k}}^{\dagger} M_{\mathbf{k}} T_{\mathbf{k}}$ is diagonal with doubly degenerate, real positive eigenvalues $\omega_{\mathbf{k},n}$,

$$T_{\mathbf{k}}^{\dagger} M_{\mathbf{k}} T_{\mathbf{k}} = \Omega_{\mathbf{k}},$$
$$\Omega_{\mathbf{k}} = \begin{pmatrix} \omega_{\mathbf{k}} & 0 \\ 0 & \omega_{\mathbf{k}} \end{pmatrix}, \quad \omega_{\mathbf{k}} = \begin{pmatrix} \omega_{\mathbf{k},1} & 0 & 0 \\ 0 & \omega_{\mathbf{k},2} & 0 \\ 0 & 0 & \omega_{\mathbf{k},3} \end{pmatrix}, \tag{A6}$$

and (ii)

$$Y T_{\mathbf{k}} Y T_{\mathbf{k}}^{\dagger} = \mathbb{I}, \quad Y = \begin{pmatrix} \mathbb{I} & 0 \\ 0 & -\mathbb{I} \end{pmatrix}. \tag{A7}$$

In terms of the blocks $U_{\mathbf{k}}$ and $V_{\mathbf{k}}$, this amounts to meeting the two following requirements:

$$\begin{aligned} U_{\mathbf{k}} U_{\mathbf{k}}^{\dagger} - V_{\mathbf{k}} V_{\mathbf{k}}^{\dagger} &= \mathbb{I}, \\ U_{\mathbf{k}} V_{\mathbf{k}}^{\dagger} - V_{\mathbf{k}} U_{\mathbf{k}}^{\dagger} &= 0. \end{aligned} \tag{A8}$$

This condition (A8) ensures that the Bogolyubov quasiparticles, which are linear combinations of the bosonic fields $a_{\mathbf{k},n}$ and $a_{\mathbf{k},n}^{\dagger}$, also obey bosonic statistics.

The zero-point energy per site can be expressed in terms of $T_{\mathbf{k}}$ as

$$\delta E = \frac{1}{2SN} \sum_{\mathbf{k}} \frac{1}{2} \mathrm{Tr}[T_{\mathbf{k}}^{\dagger} M_{\mathbf{k}} T_{\mathbf{k}}] - \mathrm{Tr}[\bar{\bar{A}}_{\mathbf{k}}]. \tag{A9}$$

According to Eq. (3.6), the $1/S$ correction to the magnetization, δm, is equal to minus the derivative of (A9) with respect to the magnetic field H. Given that $\mathrm{Tr}[\bar{\bar{A}}_{\mathbf{k}}] = 9J$ for both the Y and V states one obtains

$$\delta m = -\frac{1}{2SN} \sum_{\mathbf{k}} \frac{1}{2} \mathrm{Tr}\left[T_{\mathbf{k}}^{\dagger} \frac{\partial M_{\mathbf{k}}}{\partial H} T_{\mathbf{k}} + \frac{\partial T_{\mathbf{k}}^{\dagger}}{\partial H} M_{\mathbf{k}} T_{\mathbf{k}} + T_{\mathbf{k}}^{\dagger} M_{\mathbf{k}} \frac{\partial T_{\mathbf{k}}}{\partial H} \right]. \tag{A10}$$

Using Eq. (A6), the cyclic property of the trace, and the normalization condition (A8) one can show that the last two terms in (A10) vanish

$$\mathrm{Tr}\left[\frac{\partial T_{\mathbf{k}}^{\dagger}}{\partial H}M_{\mathbf{k}}T_{\mathbf{k}} + T_{\mathbf{k}}^{\dagger}M_{\mathbf{k}}\frac{\partial T_{\mathbf{k}}}{\partial H}\right]$$
$$= \mathrm{Tr}\left[\Omega_{\mathbf{k}}\left(\frac{\partial T_{\mathbf{k}}^{\dagger}}{\partial H}YT_{\mathbf{k}}Y + YT_{\mathbf{k}}^{\dagger}Y\frac{\partial T_{\mathbf{k}}}{\partial H}\right)\right]$$
$$= \mathrm{Tr}\left[\Omega_{\mathbf{k}}\frac{\partial}{\partial H}\begin{pmatrix} U_{\mathbf{k}}U_{\mathbf{k}}^{\dagger} - V_{\mathbf{k}}V_{\mathbf{k}}^{\dagger} & U_{\mathbf{k}}V_{\mathbf{k}}^{\dagger} - V_{\mathbf{k}}U_{\mathbf{k}}^{\dagger} \\ U_{\mathbf{k}}V_{\mathbf{k}}^{\dagger} - V_{\mathbf{k}}U_{\mathbf{k}}^{\dagger} & U_{\mathbf{k}}U_{\mathbf{k}}^{\dagger} - V_{\mathbf{k}}V_{\mathbf{k}}^{\dagger}\end{pmatrix}\right] = 0. \tag{A11}$$

The cancellation of the terms above, which is due to the normalization conditions of the eigenvectors of $M_{\mathbf{k}}$, is analogous to that occurring in the Hellmann-Feynman theorem. Hence the $1/S$ expression for the magnetization is given by

$$\delta m = -\frac{1}{2SN}\sum_{\mathbf{k}}\frac{1}{2}\mathrm{Tr}\left[T_{\mathbf{k}}^{\dagger}\frac{\partial M_{\mathbf{k}}}{\partial H}T_{\mathbf{k}}\right], \tag{A12}$$

which, given the block structure of $T_{\mathbf{k}}$, can be conveniently rewritten as

$$\delta m = -\frac{1}{SN}\sum_{\mathbf{k}}\mathrm{Tr}\left[\frac{\partial \bar{\bar{A}}_{\mathbf{k}}}{\partial H}V_{\mathbf{k}}V_{\mathbf{k}}^{\dagger} + \frac{\partial \bar{\bar{B}}_{\mathbf{k}}}{\partial H}U_{\mathbf{k}}V_{\mathbf{k}}^{\dagger}\right]. \tag{A13}$$

The derivative with respect to the field of the coefficients of $\bar{\bar{A}}_{\mathbf{k}}$ and $\bar{\bar{B}}_{\mathbf{k}}$ yields

$$\frac{\partial \bar{\bar{A}}_{\mathbf{k}}^{Y}}{\partial H} = \frac{\partial \bar{\bar{B}}_{\mathbf{k}}^{Y}}{\partial H} = \begin{pmatrix} 0 & -\gamma_{\mathbf{k}}^{\star}/12 & -\gamma_{\mathbf{k}}/12 \\ -\gamma_{\mathbf{k}}/12 & 0 & \gamma_{\mathbf{k}}^{\star}\cos\theta_2^{Y}/3 \\ -\gamma_{\mathbf{k}}^{\star}/12 & \gamma_{\mathbf{k}}\cos\theta_2^{Y}/3 & 0\end{pmatrix} \tag{A14}$$

for the Y state and

$$\frac{\partial \bar{\bar{A}}_{\mathbf{k}}^{V}}{\partial H} = \frac{\partial \bar{\bar{B}}_{\mathbf{k}}^{V}}{\partial H} = \frac{H}{36J}\begin{pmatrix} 0 & -\gamma_{\mathbf{k}}^{\star} & -\gamma_{\mathbf{k}} \\ -\gamma_{\mathbf{k}} & 0 & 0 \\ -\gamma_{\mathbf{k}}^{\star} & 0 & 0\end{pmatrix} \tag{A15}$$

for the V state. To make contact with the alternative method to compute the $1/S$ magnetization presented in the main text, we note that the two-body averages introduced in Eq. (4.3) are given by the following Brillouin zone integrals:

$$n_i = \langle a_{\mathbf{R},i}^{\dagger}a_{\mathbf{R},i}\rangle = \frac{3}{N}\sum_{\mathbf{k}}(V_{\mathbf{k}}V_{\mathbf{k}}^{\dagger})_{i,i},$$
$$m_{ij} = \langle a_{\mathbf{R},i}^{\dagger}a_{\mathbf{R}',j}\rangle = \frac{1}{N}\sum_{\mathbf{k}}\gamma_{\mathbf{k}}(V_{\mathbf{k}}V_{\mathbf{k}}^{\dagger})_{i,j}, \tag{A16}$$
$$\Delta_{ij} = \langle a_{\mathbf{R},i}a_{\mathbf{R}',j}\rangle = \frac{1}{N}\sum_{\mathbf{k}}\gamma_{\mathbf{k}}(U_{\mathbf{k}}V_{\mathbf{k}}^{\dagger})_{i,j},$$

with the sites $(\mathbf{R},i)$ and $(\mathbf{R}',j)$ being nearest neighbors. The field dependence of the averages n_i,m_{ij}, and Δ_{ij} is reported in Fig. 7. The symmetries of the Y and V structures yield $n_2 = n_3$, $m_{12} = m_{13}$, and $\Delta_{12} = \Delta_{13}$.

Injecting Eqs. (A14), (A15), and (A16) into Eq. (A13), one recovers the $1/S$ contribution to the magnetization presented

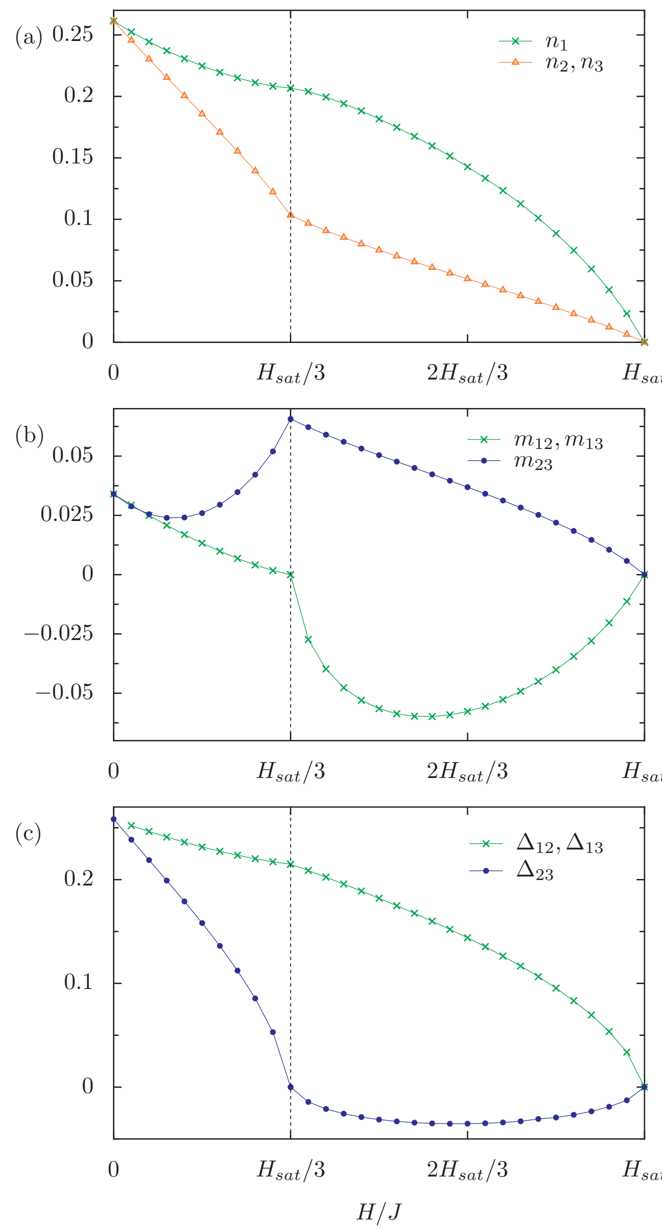

FIG. 7. Plot, as a function of the magnetic field, of the average quantities (a) $n_i = \langle a_{\mathbf{R},i}^{\dagger}a_{\mathbf{R},i}\rangle$, (b) $m_{ij} = \langle a_{\mathbf{R},i}^{\dagger}a_{\mathbf{R}',j}\rangle$, and (c) $\Delta_{ij} = \langle a_{\mathbf{R},i}a_{\mathbf{R}',j}\rangle$. We note that at $H = H_{\mathrm{sat}}/3$, i.e., for the uud state, the quantum corrections to the magnetization exactly compensate, that is, $n_1 - 2n_2 = 0$.

in Eq. (4.5), which is recalled here:

$$\delta m^{Y} = \frac{1}{3S}\left[-2\cos\theta_2^{Y}(m_{23} + \Delta_{23}) + m_{21} + \Delta_{21}\right],$$
$$\delta m^{V} = -\frac{1}{S}\frac{H}{9J}(\Delta_{21} + m_{21}). \tag{A17}$$

2. Spectrum renormalization of the uud state

The three-sublattice uud structure turns out to be classically stable at $H = H_{\mathrm{sat}}/3$. Since, according to order by disorder, collinear configurations tend to have a softer spectrum, and hence a smaller zero-point energy [41,42], quantum fluctuations stabilize this uud state over a finite field range around $H_{\mathrm{sat}}/3$, leading to the $1/3$ magnetization plateau [9]. For the specific field value $H_{\mathrm{sat}}/3$, the harmonic spectrum of the $1/S$ expansion turns out to have two gapless low-energy modes and a higher-energy gapped mode. If the uud state is to be stabilized over a given field range, it should be gapped to spin excitations. Chubukov and Golosov [9] showed that treating self-consistently the higher-order terms in the spin-wave expansion yields an excitation spectrum in which the two lowest bands are gapped. For completeness we reproduce the main steps which led Chubukov and Golosov to this conclusion.

Because of collinearity, the next nonvanishing term in the $1/S$ expansion around the uud state is quartic in boson operators. Decoupling the quartic terms [of order $O(1/S^2)$] yields an effective harmonic Hamiltonian which, up to a constant, is given by

$$\mathcal{H}^{\mathrm{eff}} = N E_{\mathrm{cl}}^{\mathrm{uud}} + \frac{1}{2S}\sum_{\mathbf{k}} \mathbf{a}_{\mathbf{k}}^{\dagger}\left[M_{\mathbf{k}}^{\mathrm{uud}}(H) + \frac{1}{S}M_{\mathbf{k}}^{\mathrm{eff}}\right]\mathbf{a}_{\mathbf{k}}\ , \quad \text{(A18)}$$

where $M_{\mathbf{k}}^{\mathrm{eff}}$ has the same block structure as $M_{\mathbf{k}}$ (A1). The subblocks of $M_{\mathbf{k}}^{\mathrm{eff}}$ are denoted by $\bar{\bar{A}}_{\mathbf{k}}^{\mathrm{eff}}$ and $\bar{\bar{B}}_{\mathbf{k}}^{\mathrm{eff}}$. Their expression can be obtained by replacing in Eq. (A2) the following coefficients:

$$\begin{aligned}
A^{\mathrm{eff}} &= 6J(-\bar{n}_2 + \bar{\Delta}_{21}),\\
B^{\mathrm{eff}} &= C^{\mathrm{eff}} = 3J(-\bar{n}_1 + \bar{n}_2 + \bar{\Delta}_{21} - \bar{m}_{23}),\\
F^{\mathrm{eff}} &= J(\bar{m}_{23} - \bar{n}_2), \quad D^{\mathrm{eff}} = G^{\mathrm{eff}} = H^{\mathrm{eff}} = 0,\\
E^{\mathrm{eff}} &= I^{\mathrm{eff}} = J[-\bar{\Delta}_{21} + (\bar{n}_1 + \bar{n}_2)/2],
\end{aligned} \quad \text{(A19)}$$

where the averages n_i,m_{ij}, and Δ_{ij} have been defined in Eq. (4.3) [see Fig. 7]. The overbar specifies that the average quantities are computed for the field $H = H_{\mathrm{sat}}/3$.

The contribution of the quartic terms renormalizes the harmonic spectrum and opens two gaps at $\mathbf{k} = 0$,

$$\begin{aligned}
\omega_{\mathbf{0}}^{(1)} &\approx \frac{1}{S}\left[H - 3J\left(1 + \frac{2\bar{m}_{23} - \bar{\Delta}_{21}}{S}\right)\right] + O(S^{-3}),\\
\omega_{\mathbf{0}}^{(2)} &\approx \frac{1}{S}\left[-H + 3J\left(1 + \frac{\bar{\Delta}_{21}}{S}\right)\right] + O(S^{-3}).
\end{aligned} \quad \text{(A20)}$$

The instability of the uud structure is resolved by determining the fields at which the gaps to the first excited states close. To first order in $1/S$, the expression of the field values at which this takes place coincides with that given in Eq. (4.7). Reference [38] provides a refinement of this approach which consists of a self-consistent treatment of the decoupling of quartic terms.

APPENDIX B: VARIATIONAL ENERGY ENVELOPE

In this Appendix we briefly mention some details of the calculation leading to the construction of a new energy curve which supports the $1/3$ magnetization plateau in the triangular-lattice Heisenberg antiferromagnet. Let us first introduce the following notations to specify the different terms entering Eq. (5.2):

$$\langle\phi_0|\mathcal{H}(H_0)|\phi_0\rangle = E_{cl}(H_0) + \frac{1}{S}\delta E(H_0), \quad \text{(B1)}$$

where $E_{cl}(H_0)$ is the classical energy at $H = H_0$ and $\delta E(H_0)/S$ is the $1/S$ correction to it. For states different from the uud structure, the magnetization, correct to order $1/S$, is obtained by deriving (B1) with respect to the field H_0,

$$\begin{aligned}
m(H_0) &= -\frac{\partial}{\partial H_0}\left(E_{cl}(H_0) + \frac{1}{S}\delta E(H_0)\right)\\
&= m_{cl}(H_0) + \frac{1}{S}\delta m(H_0),
\end{aligned} \quad \text{(B2)}$$

where $m_{cl}(H_0)$ is the classical magnetization and $\delta m(H_0)/S$ is the $1/S$ correction to it. Note that Eq. (B2) is meaningless at $H_{\mathrm{sat}}/3$. In fact, for this value of the field the harmonic energy presents a cusp, and its derivative is not well defined.

The new energy curve which is proposed [Eq. (5.3)] consists of the lower envelope of all the energies defined in Eq. (5.2). As mentioned in the main text, to compare the energies of states to order $1/S$ only the classical contribution to $\langle\phi_0|\sum_i S_i^z/S|\phi_0\rangle$ should be retained. Thus the minimization of (5.3) with respect to H_0 (again for $H_0 \neq H_{\mathrm{sat}}/3$) gives

$$\begin{aligned}
&\frac{\partial}{\partial H_0}\left(E_{cl}(H_0) + \frac{1}{S}\delta E(H_0)\right)\\
&\quad + \left(1 - (H - H_0)\frac{\partial}{\partial H_0}\right)m_{cl}(H_0) = 0\\
&\Rightarrow (H - H_0) = -\frac{1}{S}\delta m(H_0)\left(\frac{\partial m_{cl}(H_0)}{\partial H_0}\right)^{-1}\\
&\Rightarrow (H - H_0) = -\frac{1}{S}\frac{\delta m(H_0)}{\chi_{cl}},
\end{aligned} \quad \text{(B3)}$$

where we have introduced the classical susceptibility $\chi_{cl} = \partial m(H)_{cl}/\partial H$ (note that χ_{cl} is a constant since the classical magnetization depends linearly on the magnetic field). Equation (B3) establishes that the difference $H - H_0$ which minimizes (5.3) behaves as $1/S$. Retaining the $1/S$ corrections of $\langle\phi_0|\sum_i S_i^z/S|\phi_0\rangle$ in the calculation would have produced a $1/S^2$ correction to (B3).

Next, we will show that, away from the plateau, the magnetization defined as the derivative with respect to the field of the new energy envelope $\tilde{E}(H)$ differs from the $1/S$ magnetization (4.5) only by terms of order $O(1/S^2)$. For this purpose, let us compute

$$\tilde{m}(H) = -\frac{\partial\tilde{E}[H_0(H)]}{\partial H}, \quad \text{(B4)}$$

where $\tilde{E}[H_0(H)]$ is the new energy curve, with $H_0(H)$ denoting the value of H_0 fulfilling (5.3) at a given field H. After derivation one obtains

$$\begin{aligned}
\tilde{m}(H) = &-\left[\frac{\partial}{\partial H_0}\left(E_{cl}[H_0(H)] + \frac{1}{S}\delta E[H_0(H)]\right)\frac{\partial H_0}{\partial H}\right.\\
&- \left(1 - \frac{\partial H_0}{\partial H}\right)m_{cl}[H_0(H)]\\
&\left. - (H - H_0)\frac{\partial m_{cl}[H_0(H)]}{\partial H_0}\frac{\partial H_0}{\partial H}\right]\\
= &\ m_{cl}[H_0(H)],
\end{aligned} \quad \text{(B5)}$$

where we have used the first line of Eq. (B3) to simplify the expression. Thus, in this construction, we are left with a new magnetization curve

$$\tilde{m}(H) = \chi_{cl} H_0(H). \tag{B6}$$

The minimization of (5.3) does not yield a closed form $H_0(H)$; however, starting from (B3) it is straightforward to see that in the large-S limit we have

$$H_0 = H + \frac{1}{S}\frac{\delta m(H)}{\chi_{cl}} + O(1/S^2). \tag{B7}$$

Substituting (B7) into (B6) produces the result announced earlier,

$$\begin{aligned}\tilde{m}(H) &= m_{cl}(H) + \frac{1}{S}\delta m(H) + O(1/S^2)\\ &= m(H) + O(1/S^2).\end{aligned} \tag{B8}$$

So we conclude that away from the plateau, the magnetization associated with the energy curve $\tilde{E}(H)$ differs from the $1/S$ magnetization (4.5) only by terms of order $O(1/S^2)$.

[1] *Introduction to Frustrated Magnetism*, edited by C. Lacroix, P. Mendels, and F. Mila (Springer, Berlin, 2011).

[2] K. Totsuka, Phys. Rev. B **57**, 3454 (1998).

[3] F. Mila, Eur. Phys. J. B **6**, 201 (1998).

[4] P. Corboz and F. Mila, Phys. Rev. Lett. **112**, 147203 (2014).

[5] D. C. Cabra, M. D. Grynberg, P. C. W. Holdsworth, A. Honecker, P. Pujol, J. Richter, D. Schmalfuß, and J. Schulenburg, Phys. Rev. B **71**, 144420 (2005).

[6] S. Capponi, O. Derzhko, A. Honecker, A. M. Läuchli, and J. Richter, Phys. Rev. B **88**, 144416 (2013).

[7] S. Nishimoto, N. Shibata, and C. Hotta, Nat. Commun. **4**, 2287 (2013).

[8] H. Kawamura, J. Phys. Soc. Jpn. **53**, 2452 (1984).

[9] A. V. Chubukov and D. A. Golosov, J. Phys. Condens. Matter **3**, 69 (1991).

[10] M. E. Zhitomirsky, A. Honecker, and O. A. Petrenko, Phys. Rev. Lett. **85**, 3269 (2000).

[11] K. Penc, N. Shannon, and H. Shiba, Phys. Rev. Lett. **93**, 197203 (2004).

[12] T. Coletta, M. E. Zhitomirsky, and F. Mila, Phys. Rev. B **87**, 060407 (2013).

[13] A. Honecker, J. Schulenburg, and J. Richter, J. Phys. Condens. Matter **16**, S749 (2004).

[14] Y. Shirata, H. Tanaka, T. Ono, A. Matsuo, K. Kindo, and H. Nakano, J. Phys. Soc. Jpn. **80**, 093702 (2011).

[15] J. Richter, O. Götze, R. Zinke, D. J. J. Farnell, and H. Tanaka, J. Phys. Soc. Jpn. **82**, 015002 (2013).

[16] D. J. J. Farnell, R. Zinke, J. Schulenburg, and J. Richter, J. Phys. Condens. Matter **21**, 406002 (2009).

[17] T. Ono, H. Tanaka, H. Aruga Katori, F. Ishikawa, H. Mitamura, and T. Goto, Phys. Rev. B **67**, 104431 (2003).

[18] T. Ono, H. Tanaka, O. Kolomiyets, H. Mitamura, T. Goto, K. Nakajima, A. Oosawa, Y. Koike, K. Kakurai, J. Klenke *et al.*, J. Phys. Condens. Matter **16**, S773 (2004).

[19] H. Tsujii, C. R. Rotundu, T. Ono, H. Tanaka, B. Andraka, K. Ingersent, and Y. Takano, Phys. Rev. B **76**, 060406 (2007).

[20] N. A. Fortune, S. T. Hannahs, Y. Yoshida, T. E. Sherline, T. Ono, H. Tanaka, and Y. Takano, Phys. Rev. Lett. **102**, 257201 (2009).

[21] Y. Shirata, H. Tanaka, A. Matsuo, and K. Kindo, Phys. Rev. Lett. **108**, 057205 (2012).

[22] T. Susuki, N. Kurita, T. Tanaka, H. Nojiri, A. Matsuo, K. Kindo, and H. Tanaka, Phys. Rev. Lett. **110**, 267201 (2013).

[23] L. E. Svistov, A. I. Smirnov, L. A. Prozorova, O. A. Petrenko, L. N. Demianets, and A. Y. Shapiro, Phys. Rev. B **67**, 094434 (2003).

[24] L. E. Svistov, A. I. Smirnov, L. A. Prozorova, O. A. Petrenko, A. Micheler, N. Büttgen, A. Y. Shapiro, and L. N. Demianets, Phys. Rev. B **74**, 024412 (2006).

[25] A. I. Smirnov, H. Yashiro, S. Kimura, M. Hagiwara, Y. Narumi, K. Kindo, A. Kikkawa, K. Katsumata, A. Y. Shapiro, and L. N. Demianets, Phys. Rev. B **75**, 134412 (2007).

[26] M. Kenzelmann, G. Lawes, A. B. Harris, G. Gasparovic, C. Broholm, A. P. Ramirez, G. A. Jorge, M. Jaime, S. Park, Q. Huang *et al.*, Phys. Rev. Lett. **98**, 267205 (2007).

[27] J. S. White, C. Niedermayer, G. Gasparovic, C. Broholm, J. M. S. Park, A. Y. Shapiro, L. A. Demianets, and M. Kenzelmann, Phys. Rev. B **88**, 060409 (2013).

[28] M. E. Zhitomirsky and T. Nikuni, Phys. Rev. B **57**, 5013 (1998).

[29] The choice of renormalizing the bilinear spin coupling by S^2 and the magnetic field by S formally allows us to replace the quantum spin operators $\mathbf{S}_i/S$ by three-dimensional classical vectors of norm 1 in the $S \to \infty$ limit. Furthermore, this choice leads to a simple and transparent dependence on $1/S$ of the different terms of the spin-wave expansion.

[30] T. Holstein and H. Primakoff, Phys. Rev. **58**, 1098 (1940).

[31] T. Nikuni and H. Shiba, J. Phys. Soc. Jpn. **62**, 3268 (1993).

[32] H. Hellmann, *Einführung in die Quantenchemie* (Leipzig, Franz Deuticke, 1937).

[33] R. P. Feynman, Phys. Rev. **56**, 340 (1939).

[34] A. Jacobs, T. Nikuni, and H. Shiba, J. Phys. Soc. Jpn. **62**, 4066 (1993).

[35] The overbar is used to emphasize that the averages $\langle a^\dagger_{\mathbf{R},i} a_{\mathbf{R},i}\rangle$, $\langle a^\dagger_{\mathbf{R},i} a_{\mathbf{R}',j}\rangle$, and $\langle a_{\mathbf{R},i} a_{\mathbf{R}',j}\rangle$ are computed at $H = H_{\rm sat}/3 = 3J$. The bar is omitted when averages are computed at different fields.

[36] D. Yamamoto, G. Marmorini, and I. Danshita, Phys. Rev. Lett. **112**, 127203 (2014); D. Sellmann, X. F. Zhang, and S. Eggert, Phys. Rev. B **91**, 081104 (2015).

[37] D. Yamamoto, G. Marmorini, and I. Danshita, Phys. Rev. Lett. **114**, 027201 (2015).

[38] J. Takano, H. Tsunetsugu, and M. E. Zhitomirsky, J. Phys. Conf. Ser. **320**, 012011 (2011).

[39] S. Gluzman, Z. Phys. B **90**, 313 (1993).

[40] D. S. Fisher and P. C. Hohenberg, Phys. Rev. B **37**, 4936 (1988).

[41] E. F. Shender, Zh. Eksp. Teor. Fiz. **83**, 326 (1982) [Sov. Phys. JETP **56**, 178 (1982)].

[42] C. L. Henley, Phys. Rev. Lett. **62**, 2056 (1989).