Qusiclasical Hamiltonians for large-spin systems

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We extend and apply a previously developed method for a semiclassical treatment of a system with large spin $S$. A multisite Heisenberg Hamiltonian is transformed into an effective classical Hamilton function which can be treated by standard methods for classical systems. Quantum effects enter in form of multispin interactions in the Hamilton function. The latter is written in the form of an expansion in powers of $J/(TS)$, where $J$ is the coupling constant. Main ingredients of our method are spin coherent states and cumulants. Rules and diagrams are derived for computing cumulants of groups of operators entering the Hamiltonian. The theory is illustrated by calculating the quantum corrections to the free energy of a Heisenberg chain which were previously computed by a Wigner-Kirkwood expansion.

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I. INTRODUCTION

Quantum effects in systems with localized spins depend on the size of the spin $S$. They are strongest for $S = 1/2$ while systems behave classically in the limit $S \to \infty$. There are, however, numerous compounds having intermediate or high spin values, which we would like to treat semiclassically, i.e., classically with quantum corrections intermediate or high spin values, which we would like to treat semiclassically, i.e., classically with quantum corrections expanded in powers of $1/S$. Different proposals have been made in the past how this can be achieved. We have recently suggested a method by which we can derive effective classical Hamilton functions for spin Hamiltonians by using spin coherent states and cumulants. The quantum partition function is thereby replaced by a classical one, and the trace over a complete set of quantum states is replaced by an integration over the classical spin vectors. The mathematical structure of these two objects is quite different, and the transition from one to the other was investigated by Lieb who obtained upper and lower bounds for the quantum partition function in order to calculate the free energy and other thermodynamic properties of a spin chain to order $1/S^2$. These results were previously computed by a Wigner-Kirkwood expansion without using effective classical Hamiltonians.

II. SPIN COHERENT STATES AND CUMULANTS

To achieve our goal, we use two theoretical tools. The first one is the coherent-state representation of quantum-statistical averages. On each lattice site $i$, we introduce spin coherent states $|n_i\rangle$, i.e., states with the maximal projection on the axis pointing in the direction of the unit vector $n_i$. The direct product of these states $\{|\{n_i\}\rangle}$ approaches the “classical” state of the spin system in the limit $S \to \infty$. On the other hand, the basis of coherent states is complete (and even overcomplete), so that it contains all quantum states. The quantum-statistical averages of the system can be rewritten in the coherent-state basis with the help of the unity operator

$$1 = \frac{2S+1}{4\pi} \int dn \langle n|n\rangle.$$

For a single-spin system, the trace of an operator $\hat{A}$ over any complete orthonormal basis $|m\rangle$ becomes

$$\text{tr} \hat{A} = \sum_m \langle m|\hat{A}|m\rangle = \frac{2S+1}{4\pi} \int dn \sum_m \langle m|\hat{A}|n\rangle \langle n|m\rangle
\quad= \frac{2S+1}{4\pi} \int dn \langle n|\hat{A}|n\rangle.$$
Therefore the partition function for a many-spin quantum Hamiltonian $\hat{H}$ can be written as

$$Z = \left( \frac{2S+1}{4\pi} \right)^N \int \prod_{i=1}^{N} \left\{ \{n_i\} | \exp(-\beta \hat{H}) | \{n_i\} \right\},$$  

where $\beta \equiv 1/T$. It has the same form as the one for classical systems, provided one defines the effective classical Hamilton function $H$ by the relation

$$\beta H = -\ln\left\{ \{n_i\} | \exp(-\beta \hat{H}) | \{n_i\} \right\} = \langle \{n_i\} | 1 - \exp(-\beta \hat{H}) | \{n_i\} \rangle^c$$

(4)

Here the superscript $c$ denotes the cumulant of a matrix element (see below). Cumulants constitute the second theoretical tool used in this paper.

The above formula is exact for any quantum spin system. In the limit $S \to \infty$, $H$ reduces to the usual classical Hamiltonian. A mathematically rigorous treatment of this limiting transition can be found in Lieb’s paper, Ref. 3. Here we will obtain quantum corrections to the classical Hamiltonian using a systematic $1/S$ expansion for $S \gg 1$. In the same context, a technique using partial differential equations for the density matrix was applied in Ref. 4. But an explicit derivative-free expression for the effective classical Hamiltonian was obtained in Ref. 4 only for the spin-chain model under the condition that neighbouring spins are nearly colinear. The method presented here does not use derivatives and yields explicit results for any lattice.

The effective Hamilton function is obtained in form of a cumulant expansion by expanding in Eq. (4) the operator $\exp(-\beta \hat{H})$. It is worth noting a basic relation between semiclassical theory. A characteristic property of quantum mechanics is that the expectation value of a product $A_1 A_2$ of two observables with respect to a quantum state $|\psi\rangle$ is generally distinct from the product of the expectation values of $A_1$ and $A_2$. The difference is just the cumulant, i.e.,

$$\langle A_1 A_2 \rangle^c = \langle A_1 A_2 \rangle - \langle A_1 \rangle \langle A_2 \rangle \neq 0.$$  

(5)

In the classical limit this difference vanishes. Therefore we expect that a theory formulated in terms of cumulants is particularly suitable for a semiclassical expansion.

Let us recall the definition and basic properties of cumulants. Averages or matrix elements $\langle \ldots \rangle$ can be expressed through cumulants $\langle \ldots \rangle^c$ as follows

$$\langle A \rangle = \langle A \rangle^c, \quad \langle A_1 A_2 \rangle = \langle A_1 A_2 \rangle^c + \langle A_1 \rangle \langle A_2 \rangle,$$

$$\langle A_1 A_2 A_3 \rangle = \langle A_1 A_2 A_3 \rangle^c + \langle A_1 \rangle \langle A_2 A_3 \rangle^c + \langle A_2 \rangle \langle A_1 A_3 \rangle^c + \langle A_3 \rangle \langle A_1 A_2 \rangle^c + \langle A_1 \rangle \langle A_2 \rangle \langle A_3 \rangle,$$  

(6)

etc., where $A_i$ are classical stochastic variables or quantum-mechanical operators. The averaging above is performed over a classical distribution function or weighed over quantum states. A more detailed discussion of cumulants can be found in refs. 3, 4. Cumulants can be obtained by differentiation of a generating function, i.e., from

$$\langle A_1^{k_1} \ldots A_n^{k_n} \rangle^c = \frac{\partial^{k_1}}{\partial \lambda_1^{k_1}} \ldots \frac{\partial^{k_n}}{\partial \lambda_n^{k_n}} \ln \langle e^{\lambda_1 A_1} \ldots e^{\lambda_n A_n} \rangle$$  

(7)

at $\lambda_1 = \ldots = \lambda_n = 0$, in contrast to averages $\langle \ldots \rangle$, which are given by a similar expression without logarithm. By multiplying by $\prod_{i=1}^{n} \lambda_i^{k_i}/k_i!$ and summing over all $k_i = 0, \ldots, \infty$ without the term $k_1 = \ldots = k_n = 0$ one obtains the identity

$$\langle e^{\lambda_1 A_1} \ldots e^{\lambda_n A_n} - 1 \rangle = \ln \langle e^{\lambda_1 A_1} \ldots e^{\lambda_n A_n} \rangle.$$  

(8)

The second line of Eq. (4) is a particular case of this formula.

Let us consider cumulants of spin operators with respect to spin coherent states. These cumulants have an especially simple form when the spin-operator combination of spin operators. That is, the number of nonvanishing cumulants is that the expectation value of a product $S_z$ (axis $z$ along $\mathbf{n}$) and $S_\pm = S_x \pm i S_y$ are used. A cumulant vanishes if it is of the form $\langle \ldots S_z \rangle^c$, $\langle \ldots S_+ S_- \rangle^c$, $\langle \ldots S_z \rangle^c$, etc., where $\ldots$ stands for any combination of spin operators. That is, the number of $S_+$ and $S_-$ in the cumulant must be balanced in order to give a nonzero result. If, however, this balance is achieved already within a subgroup of operators on the right or left side of the operator list, the cumulant vanishes, too. Non-vanishing cumulants are

$$\langle S_z \rangle = S, \quad \langle \ldots S_z^n S_\pm^n \rangle^c = 2S(-1)^n,$$

$$\langle S_+ S_+ S_+ S_+ S_- S_- S_- S_- \rangle^c = -4S(-1)^{n+2}(-2)^m,$$

$$\langle S_+ S_+ S_+ S_+ S_- S_- S_- S_- \rangle^c = 3(S_+ S_+ S_+ S_+ S_- S_- S_- S_-) = 24S$$

(9)

etc. The above results can be obtained recurrently, using Eq. (4) and the commutation relations $[S_z, S_\pm] = -S_\pm$ and $[S_\pm, S_\pm] = 2S_z$, which remain valid inside cumulants. For scaled spins $S/S$, each nonvanishing cumulant containing $n$ spin operators scales like $1/S^{n-1}$.

An expansion of $H$ in Eq. (4) in powers of $1/S$ is obtained from the Taylor series

$$\beta H = (1 - \exp(-\beta \hat{H}))^c$$

$$= \beta \langle \hat{H} \rangle^c - \frac{\beta^2}{2!} \langle \hat{H} \hat{H} \rangle^c + \frac{\beta^3}{3!} \langle \hat{H} \hat{H} \hat{H} \rangle^c + \ldots,$$  

(10)

where we have introduced the shorthand notation $\langle \ldots \rangle \equiv \langle \{n_i\} \ldots \{n_i\} \rangle$. Here the first term on the right-hand side is the classical energy of the spin. As seen from Eq. (10), increasing powers of $1/S$ appear in each order of the expansion. In fact, Eq. (10) is an expansion in powers of $J/(S \lambda)$ and it breaks down at low temperatures. For $S \gg 1$, the range of convergence is much larger than that of the high-temperature series expansion. For the calculation of thermodynamic quantities one can further expand $\exp(-\beta \hat{H})$ in powers of $1/S$. In that case the statistical weights of different spin orientations are described by a purely classical Hamiltonian, whereas quantum effects manifest themselves in corrections to the density of states.
III. THE CUMULANT EXPANSION

Let us consider a spin Hamiltonian of the Heisenberg form

\[ \hat{H} = - \sum_i H_i \cdot S_i - \frac{1}{2} \sum_{ij} J_{ij} S_i \cdot S_j. \]  \hspace{1cm} (11)

To implement the cumulant expansion of Eq. (11), it is convenient to express the spin operator on each site \( i \) in the coordinate system with the \( z \) axis along the coherent-state vector \( n_i \equiv n_{iz} \)

\[
S_i = \sum_{\alpha_i} n_{i\alpha} S_{i\alpha}, \quad \pm = (n_x \mp i n_y)/2, \hspace{1cm} (12)
\]

where \( n_x \) and \( n_y \) are appropriate transverse basis vectors. Insertion into Eq. (11) leads to expressions of the type

\[-\frac{\beta^2}{2!^2} \sum_{i'j'} J_{i'j'} \sum_{\alpha_j,\alpha_j} \langle n_{i\alpha_j}, n_{i'\alpha_j} \rangle \langle n_{j\alpha_j}, n_{j'\alpha_j} \rangle \times (S_{i\alpha_j} S_{i'\alpha_j} S_{j'\alpha_j})^c, \hspace{1cm} (13)\]

as illustrated by the second-order pure-exchange term. The brackets inside the cumulant imply that this cumulant is defined with respect to the two pairs of the spin-operator components and not with respect to four single operators. To calculate such cumulants of composite operators, it is convenient to (i) express them through ordinary matrix elements [see, e.g., Eq. (5)] and (ii) express matrix elements through cumulants of single spin-operator components [see Eqs. (6)]. The latter are nonzero only if all the operators in the cumulant belong to the same site in which case they are readily given by Eqs. (6). One can see that in several lowest orders of the cumulant expansion, the summation over the spin-component indices \( \alpha \) in Eq. (13) reduces to a single realization which gives a nonzero result. The result of such a procedure applied to Eq. (13) is the following:

\[ \frac{1}{2^2} \langle (A_i A_{i'}) (A_j A_{j'}) \rangle^c = \langle A_i \rangle \langle A_{i'} \rangle^c \langle A_j \rangle \langle A_{j'} \rangle^c + \frac{1}{2} \langle A_i \rangle^c \langle A_{i'} \rangle^c \langle A_j \rangle \langle A_{j'} \rangle^c, \hspace{1cm} (14)\]

where \( A_i \equiv S_{i\alpha_i} \). Hereby we have taken into account that operators \( A_i \) and \( A_{i'} \) belong to different sites so that terms of the type \( \langle A_i A_{i'} \rangle^c \) vanish. On the rhs of this expression, one could add terms differing by permutations of the indices \( i, i' \) and/or \( j, j' \). These terms make the same contributions to Eq. (13) as those present in Eq. (13). Instead of writing them explicitly, they lead to the prefactor \( 1/2^2 \) in Eq. (14). Generally, the possibility of permuting spin operators connected by the exchange interaction effectively cancels the coefficient \( 1/2 \) in Eq. (14). The factor \( 1/2 \) in front of the last term of Eq. (14) appears because of the permutation of both \( i, i' \) and \( j, j' \) which does not generate a new term of this type. For the cumulant with three groups of spin operators one obtains

\[ \frac{1}{2^3} \langle (A_i A_{i'}) (A_j A_{j'}) (A_k A_{k'}) \rangle^c = \langle A_i \rangle \langle A_{i'} \rangle^c \langle A_j \rangle \langle A_{j'} \rangle^c \langle A_k \rangle \langle A_{k'} \rangle^c \]

\[ + \langle A_i \rangle \langle A_{i'} \rangle^c \langle A_j \rangle \langle A_{j'} \rangle^c \langle A_k \rangle \langle A_{k'} \rangle^c \]

\[ + \langle A_i \rangle \langle A_{i'} \rangle^c \langle A_j \rangle^c \langle A_{j'} \rangle \langle A_k \rangle \langle A_{k'} \rangle \]

\[ + \langle A_i \rangle \langle A_{i'} \rangle^c \langle A_j \rangle^c \langle A_{j'} \rangle \langle A_k \rangle^c \langle A_{k'} \rangle \]

\[ + \frac{1}{2} \langle A_i \rangle^c \langle A_{i'} \rangle^c \langle A_j \rangle^c \langle A_{j'} \rangle \langle A_k \rangle \langle A_{k'} \rangle^c \hspace{1cm} (15)\]

The rhs of Eqs. (14) and (15) are constructed according to a principle which can be formulated in a diagrammatic language: The operators \( A \) are either contracted into cumulants or connected by the interaction lines. Similarly to other diagram techniques, there are no terms consisting of disconnected parts, e.g., there is no term \( \langle A_i \rangle \langle A_{i'} \rangle \langle A_j \rangle \langle A_{j'} \rangle \) in Eq. (14). Note that the order of the operators in the cumulants on the rhs of Eqs. (14) and (15) is the same as on the left-hand side (lhs). This fact enables us to write down immediately equations of the form of Eqs. (14) or (15), without explicitly performing steps (i) and (ii). The diagrammatic representation of the cumulant expansion of Eq. (14) for the quantum Heisenberg magnet in the zero-field case is shown in Fig. 1.

![Diagram 0](image1)

Diagram 0 is the first order of the cumulant expansion in \( \hat{H} \). It will be shown below that this diagram yields the zeroth order in 1/S for the Hamilton function \( \mathcal{H} \), i.e., the classical Hamilton function \( \mathcal{H}^{(0)} \). Diagrams 1a and 1b correspond to the two terms in Eq. (14). Diagram 2a represent the three first terms in Eq. (15). These terms have the same topology and differ only by the order of spin operators. This reflects the quantum nature of the latter. Diagram 2b corresponds to the fourth term in Eq. (14). Diagram 2c represent the fifth, sixth, and seventh terms in Eq. (15). Diagrams 2d and 2e represent the two last terms, respectively.

Cumulants corresponding to the mixed field-exchange terms in the cumulant expansion can be treated in a similar manner. One obtains

\[ \frac{1}{2} \langle (A_i A_{i'}) A_j \rangle^c = \langle A_i \rangle \langle A_{i'} \rangle^c \langle A_j \rangle \]

\[ + \frac{1}{2} \langle A_i \rangle^c \langle A_{i'} \rangle^c \langle A_j \rangle \]
terms in the cumulant expansion of Eq. (10). The term
the help of Eqs. (14), (16), and (9) one obtains
Eqs. (16), in particular, the diagram corresponding to the
\( H_{2} \equiv \langle 1 \rangle \)
functions \( \Phi \) in Eq. (21) we find
form of Eq. (15), the 2-spin interactions appear.
One can see that the effective classical Hamilton func-
tion corresponding to the Heisenberg quantum Hamilton-
ian, Eq. (14), is of a non-Heisenberg form. In particular,
many-spin interactions appear.

Let us now proceed to the third order terms of the
cumulant expansion. At first we consider the zero-field
case. Equation (13) generates terms of different topology
in \( \mathcal{H}^{(2)} \) (see Fig. 3)

\[
\mathcal{H}^{(1)} = -\frac{\beta}{4S} \sum_{i} [h_i \times n_i]^2 \\
- \frac{\beta}{4S} \sum_{ij} \tilde{J}_{ij} [(h_i \cdot n_j) - (h_j \cdot n_i)(n_i \cdot n_j)] \\
- \frac{\beta}{4S} \sum_{ij} \tilde{J}_{ij} (n_i \cdot n_j) - (n_i \cdot n_j)(n_i \cdot n_j)] \\
- \frac{\beta}{16S^2} \sum_{ij} \tilde{J}_{ij}^2 [1 - (n_i \cdot n_j)]^2. 
\]

IV. THE EFFECTIVE HAMILTONIAN

Let us now proceed to the calculation of the different
terms in the cumulant expansion of Eq. (13). The term
of the first order in \( H \) yields the classical Hamiltonian

\[
\mathcal{H}^{(0)} = -\sum_{i} h_i \cdot n_i - \frac{1}{2} \sum_{ij} \tilde{J}_{ij} n_i \cdot n_j, 
\]

which is the zeroth order of the 1/S expansion. Here

\[
h_i \equiv SH_i, \quad \tilde{J}_{ij} \equiv S^2 J_{ij} 
\]

are the reduced magnetic field and the exchange interaction,
respectively. In second and higher orders in \( \tilde{H} \) of the
cumulant expansion, 1/S corrections appear. With the help of Eqs. (14), (16), and (18) one obtains

\[
\mathcal{H}^{(1)} = -\frac{\beta}{S} \sum_{i} (h_i \cdot n_{i+} + h_i \cdot n_{i-}) \\
- \frac{\beta}{S} \sum_{ij} \tilde{J}_{ij} [(h_i \cdot n_{i+} + h_i \cdot n_{i-}) + (h_j \cdot n_{i+} + h_j \cdot n_{i-})] \\
- \frac{\beta}{S} \sum_{ij} \tilde{J}_{ij} \tilde{J}_{jl} (n_i \cdot n_{j+} + n_i \cdot n_{l+}) \\
- \frac{\beta}{S^2} \sum_{ij} \tilde{J}_{ij}^2 (n_{i+} \cdot n_{j+} + n_{i-} \cdot n_{j-}). 
\]

In this expression, as well as in all other expressions of
this type, the transverse components of the coherent-
state vectors, \( n_{\pm} \), can be expressed in terms of \( n \) after
applying some vector algebra (for details see Appendix).
This is not surprising since \( n \) is the only vector specifying
the spin coherent state. Thus one can rewrite \( \mathcal{H}^{(1)} \) in the
form

\[
\mathcal{H}^{(1)} = -\frac{\beta}{4S} \sum_{i} [h_i \times n_i]^2 \\
- \frac{\beta}{4S} \sum_{ij} \tilde{J}_{ij} [(h_i \cdot n_j) - (h_j \cdot n_i)(n_i \cdot n_j)] \\
- \frac{\beta}{4S} \sum_{ij} \tilde{J}_{ij} (n_i \cdot n_j) - (n_i \cdot n_j)(n_i \cdot n_j)] \\
- \frac{\beta}{16S^2} \sum_{ij} \tilde{J}_{ij}^2 [1 - (n_i \cdot n_j)]^2. 
\]
\[ \Phi_{2b} = -4(n_i \cdot n_{a+})(n_j \cdot n_a)(n_i \cdot n_a) - \frac{1}{3}(n_i \cdot n_a)(n_j \cdot n_a) + (n_j \cdot n_a)(n_i \cdot n_i) + (n_i \cdot n_a)(n_i \cdot n_j) \]  
= \left[ \sum_{ij} (n_i \cdot h_i)(n_j \cdot h_j) \right]^2  
= \left( \sum_{ij} \Phi_{2h}^2 \right) \left( \sum_{ij} \Phi_{2h,b}^2 \right)  
= \left( \sum_{ij} \Phi_{2h}^2 \right) \left( \sum_{ij} \Phi_{2h,b}^2 \right)  
\]

V. PHYSICAL QUANTITIES

Quantum-statistical averages of operators describing various physical quantities can be obtained by differentiation of the partition function \( Z \) or its logarithm with respect to appropriate parameters. For the internal energy \( U = \langle H \rangle \) one obtains

\[ U = -\partial \ln Z / \partial \beta = \langle H^* \rangle, \]

where \( \langle \ldots \rangle \) denotes a classical thermal average and
\[ \mathcal{H}^* = \partial(\beta \mathcal{H})/\partial \beta = \mathcal{H}^{(0)} + 2\mathcal{H}^{(1)} + 3\mathcal{H}^{(2)} + \ldots \] (34)

differs from \( \mathcal{H} = \mathcal{H}^{(0)} + \mathcal{H}^{(1)} + \mathcal{H}^{(2)} + \ldots \). The scaled magnetization per site, \( m \equiv \langle \langle S \rangle \rangle /(SN) \equiv \sum_i \langle \langle S_i \rangle \rangle /SN \), is given by

\[ m = \frac{1}{N} \frac{\partial \ln Z}{\partial (\beta h)} = \frac{1}{N} \langle n^* \rangle, \quad n^* = -\frac{\partial \mathcal{H}}{\partial h}, \] (35)

where \( h \) is the homogeneous part of the magnetic field defined formally by \( h_i = h + \mathbf{h} \). Here, \( n^* \) is not just \( n = \sum_i n_i \) but contains quantum corrections from all orders of the cumulant expansion. In particular, to first order of the cumulant expansion, one obtains from Eqs. (17) and (20)

\[ n^* = \sum_i n_i + \frac{\beta}{2S} \sum_i [h_i - n_i(n_i \cdot \mathbf{h})]
+ \frac{\beta}{4S} \sum_{ij} \tilde{J}_{ij} [1 - (n_i \cdot n_j)](n_i + n_j). \] (36)

The reduced correlation function of different spin components on different lattice sites can be written as

\[ \frac{1}{S^2} \langle \langle S_{\alpha i} S_{\beta j} \rangle \rangle = \frac{1}{Z} \frac{\partial^2 Z}{\partial (\beta h_{\alpha i}) \partial (\beta h_{\beta j})} = \langle \frac{\partial \mathcal{H}}{\partial h_{\alpha i}} \frac{\partial \mathcal{H}}{\partial h_{\beta j}} - \frac{\partial^2 \mathcal{H}}{\partial h_{\alpha i} \partial h_{\beta j}} \rangle. \] (37)

One notices that in order to calculate a correlation function, it is insufficient to perform a classical thermal average of \( n_{\alpha i} n_{\beta j} \) or even of \( n^*_{\alpha i} n^*_{\beta j} \). The last term of Eq. (27) makes a contribution to third order in the cumulant expansion due to the terms of the type \( JH^2 \) [see Eq. (28)].

VI. APPLICATION TO THE SPIN CHAIN

The isotropic spin chain in zero magnetic field is a simply solvable model in the classical limit [3]. The effective quasiclassical Hamiltonian discussed in this paper can be used to analytically calculate \( 1/S \) corrections to the classical results. The \( 1/S \) expansion of the partition function has the form

\[ Z \simeq \tilde{Z}_0 \left[ 1 - \langle \beta (\mathcal{H}^{(1)} + \mathcal{H}^{(2)}) \rangle + \frac{1}{2!} \langle [\beta \mathcal{H}^{(1)}]^2 \rangle + \ldots \right], \] (38)

where the averages are performed with respect to the classical Hamiltonian \( \mathcal{H}^{(0)} \),

\[ \tilde{Z}_0 = \left( \frac{2S + 1}{4\pi} \right)^N Z_0, \] (39)

and \( Z_0 \) is the partition function of the classical system. For the open spin chain the latter is given by \( Z_0 = 4\pi (4\pi \sinh(\xi)/\xi)^{N-1} \) with \( \xi = \beta J \). To order \( 1/S^2 \), one should use for the linear \( \mathcal{H}^{(1)} \) term in Eq. (38) the third and fourth terms of Eq. (20), for the quadratic \( \mathcal{H}^{(1)} \) term the third term of Eq. (24), and for the linear \( \mathcal{H}^{(2)} \) term the first and second terms of Eq. (21). After performing thermodynamic averages one obtains

\[ \frac{\ln Z}{N} \simeq \ln(2S + 1) + \ln \left( \frac{\sinh \xi}{\xi} \right) + \frac{\beta B}{S} + \frac{5\xi^2 - 7\xi^2 B^2 + 3\xi^2 B - 9\xi B}{12S^2} + O \left( \frac{1}{S^3} \right), \] (40)

for ferro- and antiferromagnets, where \( B \equiv \coth \xi - 1/\xi \) is the Langevin function. This formula was obtained earlier [2] with the help of the Wigner-Kirkwood expansion which avoids using effective classical Hamiltonians. It strongly resembles the result for the two-spin model, Eq. (20) of Ref. [3], where the factor 7 is replaced by 6. For the energy per spin \( U = -\beta \ln Z/\beta \) one obtains

\[ \frac{U}{J} \equiv -B - \frac{1}{S} [B + \xi B'] - \frac{1}{S^2} [10\xi - (14\xi B + 9)(B + \xi B') \mp (2B + \xi B')]. \] (41)

Let us consider now the energy \( U \) per spin which follows from linear spin-wave theory

\[ U = -\tilde{J}_0 - \frac{\tilde{J}_0}{2S} \int dq \left( 1 - \tilde{\varepsilon}_q \right) + T \int dq \left( \frac{dq}{2\pi d} \exp(\tilde{\beta} \tilde{q}^2) - 1 \right), \]

\[ \tilde{\beta} \equiv \beta \tilde{J}_0/S, \quad \tilde{\varepsilon}_q \equiv (S/\tilde{J}_0) \varepsilon_q = \left\{ \frac{1}{1 - \lambda q} \right\}^F \left\{ \frac{1}{1 - \lambda q} \right\}^A F. \] (42)

Here \( \tilde{J}_0 \) is the zeroth Fourier component of \( \tilde{J}_{ij} \) and \( \lambda_q \equiv \tilde{J}_q/\tilde{J}_0 \). The first term in the expression for \( U \) is the classical ground-state energy, the second term is the quantum correction to the former, and the last term is the temperature-dependent magnon contribution. Strictly speaking, linear spin-wave theory is only applicable for higher than two dimensions, but, although, Eq. (42) remains well-defined in lower dimensions. The Haldane gap in the magnon spectrum for integer \( S \), which is not taken into account in Eq. (42), behaves as \( \exp(-S) \) and becomes negligible for large spins. Whereas \( \tilde{\varepsilon}_q \) is of order unity, the parameter \( \tilde{\beta} \) is precisely the small parameter of the cumulant expansion. If one expands \( U \) in powers of \( \tilde{\beta} \), one obtains a series which is very close to the one following from the cumulant expansion in the limit \( \xi \equiv J/T \gg 1 \). In particular, for a ferromagnetic chain in the temperature interval \( J/T \ll T \ll J \) Eq. (41) yields

\[ \frac{U}{J} \equiv -1 - \frac{1}{S} + \frac{\xi}{\xi} + \frac{\xi}{2S^2} - \frac{1}{2S^2}, \] (43)

whereas expanding Eq. (42) we obtain the same expression without the last term. In the antiferromagnetic case one obtains similar expressions with coefficients 6 and 3 instead of 2 and 2 in the denominator.

In Fig. 3 we compare the heat capacity \( C = dU/dT \) of a Heisenberg antiferromagnetic chain with \( S = 3/2 \) calculated in different orders of the cumulant expansion [see
Eq. (11) and following from the linear spin-wave theory [see Eq. (12)] with numerical result of Ref. [14], where the transfer-matrix renormalization group (TMRG) was used. Although a spin value 3/2 is not large, one notices that taking into account quantum corrections improves the behavior of the heat capacity provided the temperature is not too low. In order to achieve quantitative agreement with the numerically accurate result, one must take into account higher-order corrections in 1/S. This requires calculation of the next terms of the effective classical Hamiltonian, which can be done with the help of the diagram technique developed in Sec. II. Also on the low-temperature side the accuracy of the SWT result can be improved by taking in Eq. (12) into account 1/S corrections to the magnon spectrum.

VII. DISCUSSION

In the preceding part of the paper, we have obtained an effective classical Hamiltonian $\mathcal{H}$ for the large-spin quantum magnet described by the Heisenberg Hamiltonian $\hat{H}$, Eq. (11). $\mathcal{H}$ consists of a purely classical part and quantum corrections of different orders in 1/S. Quantum corrections have a non-Heisenberg form and their structure becomes more and more complicated with increasing order of 1/S. In particular, pair interactions in $\hat{H}$ give rise to many-spin interactions in $\mathcal{H}$.

Our effective classical Hamilton function $\mathcal{H}$ looks different from the effective classical Hamilton function obtained in Refs. [3]. The latter has the same Heisenberg form, and the quantum effects are absorbed in the factor renormalizing the exchange interaction. This factor satisfies a system of nonlinear equations. It is difficult to make a direct comparison of the two effective classical Hamilton functions because of their different structures and different ways of derivation: Our approach is perturbative in 1/S and it leads to the same results for the physical quantities as the Wigner-Kirkwood expansion, whereas the approach of Refs. [3] is nonperturbative.

The effective classical Hamilton function $\mathcal{H}$ obtained in this paper can be used to compute quantum corrections for magnetic systems with large spins and not too low temperatures. For one-dimensional models, this was done with the help of analytical methods. In two and higher dimensions, one can apply the diagram technique for classical spins (see, e.g., Ref. [13]), which should be generalized to non-Heisenberg Hamiltonians, however. For models without long-range order, such as two-dimensional ferro- and antiferromagnets, it is problematic to sum up the relevant diagrams for the corresponding classical case when the temperature is low. Then the 1/D expansion, where $D$ is the number of spin components, proves to be an efficient tool for low-dimensional classical magnets [13] [14]. Since $\mathcal{H}$ is written in terms of various scalar products, it can be easily generalized to arbitrary values of $D$ and treated with the help of a 1/D expansion.

APPENDIX: ELIMINATION OF TRANSVERSE COMPONENTS OF THE COHERENT-STATE VECTORS

There are two generic formulas for the elimination of transverse components of the coherent-state vectors. The first one,

$$\sum_{\alpha=x,y} (\mathbf{a} \cdot \mathbf{n}_\alpha)(\mathbf{n}_\alpha \cdot \mathbf{b}) = (\mathbf{a} \cdot \mathbf{b}) - (\mathbf{a} \cdot \mathbf{n})(\mathbf{n} \cdot \mathbf{b}),$$

(44)

where $\mathbf{a}$ and $\mathbf{b}$ are arbitrary vectors, follows from the definition of the scalar product $(\mathbf{a} \cdot \mathbf{b})$. The second formula is

$$(\mathbf{a} \cdot \mathbf{n}_x)(\mathbf{n}_y \cdot \mathbf{b}) - (\mathbf{a} \cdot \mathbf{n}_y)(\mathbf{n}_x \cdot \mathbf{b}) = \left((\mathbf{n}_x \times \mathbf{n}_y) \cdot (\mathbf{a} \times \mathbf{b})\right) = (\mathbf{n} \cdot (\mathbf{a} \times \mathbf{b})).$$

(45)

Combining these two formulas one obtains the relation

$$4(\mathbf{a} \cdot \mathbf{n}_\pm)(\mathbf{n}_\mp \cdot \mathbf{b}) = (\mathbf{a} \cdot \mathbf{b}) - (\mathbf{a} \cdot \mathbf{n})(\mathbf{n} \cdot \mathbf{b})$$

$$\pm i(\mathbf{n} \cdot (\mathbf{a} \times \mathbf{b})), \quad (46)$$

which is used in the main text to eliminate $\mathbf{n}_\pm$. Other useful relations are

$$\begin{align*}
(\mathbf{n}_+ \cdot \mathbf{n}_-) &= \frac{1}{4}(n_x^2 + n_y^2) = \frac{1}{2}, \\
[n_+ \times n_-] &= \frac{i}{2} \mathbf{n}, \quad [n_\pm \times \mathbf{n}] = \mp i n_\pm. \quad (47)
\end{align*}$$

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