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Quasiclassical Hamiltonians for large-spin systems

D.A. Garanin[1,a], K. Kladko[2], and P. Fulde[1]

[1] Max-Planck-Institut für Physik komplexer Systeme, Nöthnitzer Strasse 38, 01187 Dresden, Germany
[2] Condensed Matter Physics Group and Center for Nonlinear Studies, Theoretical Division, LANL, MS-B258, Los Alamos, NM 87545, USA

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Abstract. 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.

PACS. 05.30.-d Quantum statistical mechanics – 75.10.Hk Classical spin models – 75.10.Jm Quantized spin models

1 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 expanded in powers of $1/S$. Different proposals have been made in the past how this can be achieved [1,2]. 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 [3]. 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 [4] who obtained upper and lower bounds for the quantum partition function for large S. He did not consider the $1/S$ corrections to the classical partition function which are discussed here.

The previous paper [3] dealt with one- and two-spin systems and their semiclassical description when S is large. Here we extend these calculations to a many-spin system. The necessary formalism is described in Section 2. The method is applied in Section 3 to the Heisenberg Hamiltonian in an applied magnetic field first stating the rules for evaluating the required cumulants. As we will show this can be done by means of a diagrammatic technique. The explicit form of the corresponding classical Hamilton function is derived in Section 4. All terms of order $J[J/(TS)]^2/S^n$ with $n = 0, 1, 2$ are included in the effective Hamilton function. Obviously, the semiclassical description of large-spin systems breaks down in the low-temperature limit, where quantization of energy levels becomes important. To check the method, we use in Section 6 our effective Hamilton 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. Section 7 contains a discussion and the conclusions.

2 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* $|\mathbf{n}_i\rangle$, *i.e.*, states with the maximal projection on the axis pointing in the direction of the unit vector $\mathbf{n}_i$. The direct product of these states $|\{\mathbf{n}_i\}\rangle$ 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

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[a] e-mail: `garanin@mpipks-dresden.mpg.de`

with the help of the unity operator

$$\mathbf{1} = \frac{2S+1}{4\pi}\int \mathrm{d}\mathbf{n}\,|\mathbf{n}\rangle\langle\mathbf{n}|. \tag{1}$$

For a single-spin system, the trace of an operator $\hat{A}$ over any complete orthonormal basis $|m\rangle$ becomes [4]

$$\begin{aligned}\operatorname{tr}\hat{A} = \sum_m \langle m|\hat{A}|m\rangle &= \frac{2S+1}{4\pi}\int \mathrm{d}\mathbf{n}\sum_m \langle m|\hat{A}|\mathbf{n}\rangle\langle\mathbf{n}|m\rangle \\ &= \frac{2S+1}{4\pi}\int \mathrm{d}\mathbf{n}\,\langle\mathbf{n}|\hat{A}|\mathbf{n}\rangle. \end{aligned}\tag{2}$$

Therefore the partition function for a many-spin quantum Hamiltonian $\hat{H}$ can be written as

$$\mathcal{Z} = \left(\frac{2S+1}{4\pi}\right)^N \int \prod_{i=1}^{N} \mathrm{d}\mathbf{n}_i\,\langle\{\mathbf{n}_i\}|\exp(-\beta\hat{H})|\{\mathbf{n}_i\}\rangle, \tag{3}$$

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

$$\begin{aligned}\beta\mathcal{H} &= -\ln\langle\{\mathbf{n}_i\}|\exp(-\beta\hat{H})|\{\mathbf{n}_i\}\rangle \\ &= \langle\{\mathbf{n}_i\}|1-\exp(-\beta\hat{H})|\{\mathbf{n}_i\}\rangle^c.\end{aligned}\tag{4}$$

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 $S \to \infty$, $\mathcal{H}$ reduces to the usual classical Hamiltonian. A mathematically rigorous treatment of this limiting transition can be found in Lieb's paper, reference [4] 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 reference [1]. But an explicit derivative-free expression for the effective classical Hamiltonian was obtained in reference [1] only for the spin-chain model under the condition that neigbouring spins are nearly collinear. 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 equation (4) the operator $\exp(-\beta\hat{H})$. It is worth noting a basic relation between semiclassical theory and cumulants. A characteristic property of quantum mechanics is that the expectation value of a product A_1A_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.*,

$$\langle A_1A_2\rangle^c = \langle A_1A_2\rangle - \langle A_1\rangle\langle A_2\rangle \neq 0. \tag{5}$$

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

$$\begin{aligned}\langle A\rangle = \langle A\rangle^c, \quad \langle A_1A_2\rangle &= \langle A_1A_2\rangle^c + \langle A_1\rangle\langle A_2\rangle, \\ \langle A_1A_2A_3\rangle = \langle A_1A_2A_3\rangle^c &+ \langle A_1\rangle\langle A_2A_3\rangle^c + \langle A_2\rangle\langle A_1A_3\rangle^c \\ &+ \langle A_3\rangle\langle A_1A_2\rangle^c + \langle A_1\rangle\langle A_2\rangle\langle A_3\rangle,\end{aligned}\tag{6}$$

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 references [5–7]. Cumulants can be obtained by differentiation of a generating function, *i.e.*, from

$$\langle A_1^{k_1}\ldots A_n^{k_n}\rangle^c = \frac{\partial^{k_1}}{\partial\lambda_1^{k_1}}\cdots\frac{\partial^{k_n}}{\partial\lambda_n^{k_n}}\ln\langle \mathrm{e}^{\lambda_1A_1}\ldots\mathrm{e}^{\lambda_nA_n}\rangle \tag{7}$$

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

$$\langle \mathrm{e}^{\lambda_1A_1}\ldots\mathrm{e}^{\lambda_nA_n} - 1\rangle^c = \ln\langle \mathrm{e}^{\lambda_1A_1}\ldots\mathrm{e}^{\lambda_nA_n}\rangle. \tag{8}$$

The second line of equation (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 components S_z (axis z along $\mathbf{n}$) and $S_\pm = S_x \pm \mathrm{i}S_y$ are used. A cumulant vanishes if it is of the form $\langle\ldots S_+\rangle^c$, $\langle\ldots S_+S_-\rangle^c$, $\langle\ldots S_z\rangle^c$, 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

$$\begin{aligned}&\langle S_z\rangle = S, \quad \langle S_+S_z^nS_-\rangle^c = 2S(-1)^n, \\ &\langle S_+S_z^nS_+S_z^mS_-S_z^{n'}S_-\rangle^c = -4S(-1)^{n+n'}(-2)^m, \\ &\quad\langle S_+S_+S_+S_-S_-S_-\rangle^c = 3\langle S_+S_+S_-S_+S_-S_-\rangle^c = 24S,\end{aligned}\tag{9}$$

etc. The above results can be obtained recurrently, using equation (6) and the commutation relations $[S_z,S_-] = -S_-$ and $[S_+,S_-] = 2S_z$, which remain valid inside cumulants. For scaled spins $\mathbf{S}/S$, each nonvanishing cumulant containing n spin operators scales like $1/S^{n-1}$.

An expansion of $\mathcal{H}$ in equation (4) in powers of $1/S$ is obtained from the Taylor series

$$\begin{aligned}\beta\mathcal{H} &= \langle 1-\exp(-\beta\hat{H})\rangle^c \\ &= \beta\langle\hat{H}\rangle^c - \frac{\beta^2}{2!}\langle\hat{H}\hat{H}\rangle^c + \frac{\beta^3}{3!}\langle\hat{H}\hat{H}\hat{H}\rangle^c + \ldots,\end{aligned}\tag{10}$$

where we have introduced the shorthand notation $\langle \dots \rangle \equiv \langle \{\mathbf{n}_i\} | \dots | \{\mathbf{n}_i\} \rangle$. Here the first term on the right-hand side (rhs) is the classical energy of the spin. As seen from equation (9), increasing powers of $1/S$ appear in each order of the expansion. In fact, equation (10) is an expansion in powers of $J/(TS)$ 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 $\exp(-\beta\mathcal{H})$ 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.*

3 The cumulant expansion

Let us consider a spin Hamiltonian of the Heisenberg form

$$\hat{H} = -\sum_i \mathbf{H}_i \cdot \mathbf{S}_i - \frac{1}{2} \sum_{ij} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j. \tag{11}$$

To implement the cumulant expansion of equation (10), 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 $\mathbf{n}_i \equiv \mathbf{n}_{iz}$

$$\mathbf{S}_i = \sum_{\alpha_i = z,\pm} \mathbf{n}_{i\alpha_i} S_{i\alpha_i} \qquad \mathbf{n}_\pm \equiv (\mathbf{n}_x \mp \mathrm{i}\mathbf{n}_y)/2, \tag{12}$$

where $\mathbf{n}_x$ and $\mathbf{n}_y$ are appropriate transverse basis vectors. Insertion into equation (10) leads to expressions of the type

$$-\frac{\beta^2}{2!\,2^2} \sum_{ii'jj'} J_{ii'} J_{jj'} \sum_{\alpha_i \alpha_{i'} \alpha_j \alpha_{j'}} (\mathbf{n}_{i\alpha_i} \cdot \mathbf{n}_{i'\alpha_{i'}})(\mathbf{n}_{j\alpha_j} \cdot \mathbf{n}_{j'\alpha_{j'}}) \times \langle (S_{i\alpha_i} S_{i'\alpha_{i'}})(S_{j\alpha_j} S_{j'\alpha_{j'}}) \rangle^c, \tag{13}$$

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 equations (9). One can see that in several lowest orders of the cumulant expansion, the summation over the spin-component indices α in equation (13) reduces to a single realization which gives a nonzero result. The result of such a procedure applied to equation (13) is the following:

$$\frac{1}{2^2} \langle (A_i A_{i'})(A_j A_{j'}) \rangle^c = \langle A_i \rangle \langle A_{i'} A_j \rangle^c \langle A_{j'} \rangle + \frac{1}{2} \langle A_i A_j \rangle^c \langle A_{i'} A_{j'} \rangle^c, \tag{14}$$

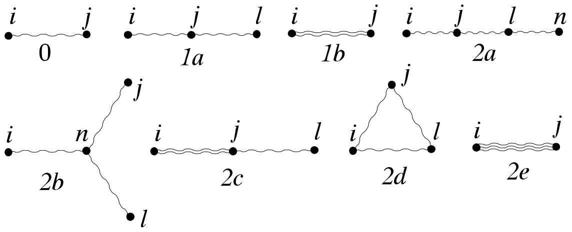

Fig. 1. Diagrammatical representation of the cumulant expansion for the quantum Heisenberg magnet. Wavy lines stand for the exchange interaction J_{ij}, small solid circles are cumulants of spin operators.

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 equation (13) as those present in equation (14); Instead of writing them explicitly, they lead to the prefactor $1/2^2$ in equation (14). Generally, the possibility of permutating spin operators connected by the exchange interaction effectively cancels the coefficient $1/2$ in equation (11). The factor $1/2$ in front of the last term of equation (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

$$\begin{aligned} \frac{1}{2^3} \langle (A_i A_{i'})(A_j A_{j'})(A_l A_{l'}) \rangle^c = {} & \langle A_i \rangle \langle A_{i'} A_j \rangle^c \langle A_{j'} A_l \rangle^c \langle A_{l'} \rangle \\ & + \langle A_i \rangle \langle A_{i'} A_l \rangle^c \langle A_j A_{l'} \rangle^c \langle A_{j'} \rangle \\ & + \langle A_j \rangle \langle A_i A_{j'} \rangle^c \langle A_{i'} A_l \rangle^c \langle A_{l'} \rangle \\ & + \langle A_i A_j A_l \rangle^c \langle A_{i'} \rangle \langle A_{j'} \rangle \langle A_{l'} \rangle \\ & + \langle A_i A_j \rangle^c \langle A_{i'} A_{j'} A_l \rangle^c \langle A_{l'} \rangle \\ & + \langle A_i A_l \rangle^c \langle A_{i'} A_j A_{l'} \rangle^c \langle A_{j'} \rangle \\ & + \langle A_j A_l \rangle^c \langle A_i A_{j'} A_{l'} \rangle^c \langle A_{i'} \rangle \\ & + \langle A_{i'} A_j \rangle^c \langle A_{j'} A_l \rangle^c \langle A_i A_{l'} \rangle^c \\ & + \frac{1}{2} \langle A_i A_j A_l \rangle^c \langle A_{i'} A_{j'} A_{l'} \rangle^c. \end{aligned} \tag{15}$$

The rhs of equations (14, 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 equation (14). Note that the order of the operators in the cumulants on the rhs of equations (14, 15) is the same as on the left-hand side (lhs). This fact enables us to write down immediately, equations of the form of equations (14) or (15), without explicitly performing steps (i) and (ii). The diagrammatic representation of the cumulant expansion of equation (10) for the quantum Heisenberg magnet in the zero-field case is shown in Figure 1. Diagram 0 is the first order of the cumulant expansion in $\hat{H}$. It will be shown below that

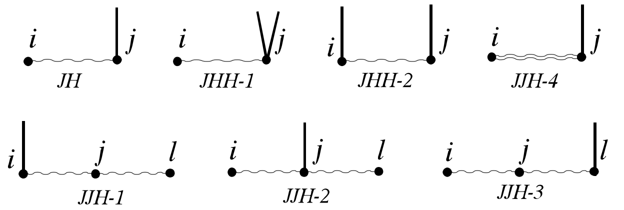

Fig. 2. The same as in Figure 1 for the mixed field-exchange terms. Straight lines represent an external magnetic field.

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 equation (14). Diagram 2a represents the first three terms in equation (15). These terms have the same topology and differ only by the order of the spin operators. This reflects its quantum nature. Diagram 2b corresponds to the fourth term in equation (15). Diagram 2c represent the fifth, sixth, and seventh terms in equation (15). Diagrams 2d and 2e represent the last two terms, respectively.

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

$$\begin{aligned}
\frac{1}{2}\langle (A_i A_{i'}) A_j\rangle^c &= \langle A_i\rangle\langle A_{i'} A_j\rangle^c \\
\frac{1}{2}\langle A_j (A_i A_{i'})\rangle^c &= \langle A_i\rangle\langle A_j A_{i'}\rangle^c \\
\frac{1}{2}\langle (A_i A_{i'}) A_j A_l\rangle^c &= \langle A_i\rangle\langle A_{i'} A_j A_l\rangle^c \\
&\quad + \langle A_i A_l\rangle^c \langle A_{i'} A_j\rangle^c \\
\frac{1}{2^2}\langle (A_i A_{i'})(A_j A_{j'}) A_l\rangle^c &= \langle A_i A_l\rangle^c \langle A_{i'} A_j\rangle^c \langle A_{j'}\rangle \\
&\quad + \langle A_i\rangle\langle A_{i'} A_j A_l\rangle^c \langle A_{j'}\rangle \\
&\quad + \langle A_i\rangle\langle A_{i'} A_j\rangle^c \langle A_{j'} A_l\rangle^c \\
&\quad + \langle A_i A_j A_l\rangle^c \langle A_{i'} A_{j'}\rangle^c, \qquad (16)
\end{aligned}$$

etc. The diagrams corresponding to the rhs of these expressions are shown in Figure 2. For the last equation of (16), in particular, the diagram corresponding to the nth term ($n = 1, 2, 3, 4$) on the rhs is labeled by $JJH\text{-}n$.

4 The effective Hamiltonian

Let us now proceed to the calculation of the different terms in the cumulant expansion of equation (10). The term of the first order in $\hat{H}$ yields the classical Hamiltonian

$$\mathcal{H}^{(0)} = -\sum_i \mathbf{h}_i \cdot \mathbf{n}_i - \frac{1}{2}\sum_{ij} \tilde{J}_{ij} \mathbf{n}_i \cdot \mathbf{n}_j, \qquad (17)$$

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

$$\mathbf{h}_i \equiv S\mathbf{H}_i, \quad \tilde{J}_{ij} \equiv S^2 J_{ij} \qquad (18)$$

are the reduced magnetic field and the exchange interaction, respectively. In second and higher orders in $\hat{H}$ of the cumulant expansion, $1/S$ corrections appear. With the help of equations (14, 16, 9) one obtains

$$\begin{aligned}
\mathcal{H}^{(1)} = &-\frac{\beta}{S}\sum_i (\mathbf{h}_i \cdot \mathbf{n}_{i+})(\mathbf{n}_{i-} \cdot \mathbf{h}_i) \\
&- \frac{\beta}{S}\sum_{ij} \tilde{J}_{ij}[(\mathbf{h}_i \cdot \mathbf{n}_{i+})(\mathbf{n}_{i-} \cdot \mathbf{n}_j) + (\mathbf{h}_i \cdot \mathbf{n}_{i-})(\mathbf{n}_{i+} \cdot \mathbf{n}_j)] \\
&- \frac{\beta}{S}\sum_{ijl} \tilde{J}_{ij}\tilde{J}_{jl}(\mathbf{n}_i \cdot \mathbf{n}_{j+})(\mathbf{n}_{j-} \cdot \mathbf{n}_l) \\
&- \frac{\beta}{S^2}\sum_{ij} \tilde{J}_{ij}^2 (\mathbf{n}_{i+} \cdot \mathbf{n}_{j+})(\mathbf{n}_{i-} \cdot \mathbf{n}_{j-}). \qquad (19)
\end{aligned}$$

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

$$\begin{aligned}
\mathcal{H}^{(1)} = &-\frac{\beta}{4S}\sum_i [\mathbf{h}_i \times \mathbf{n}_i]^2 \\
&- \frac{\beta}{4S}\sum_{ij} \tilde{J}_{ij}[(\mathbf{h}_i \cdot \mathbf{n}_j) - (\mathbf{h}_i \cdot \mathbf{n}_i)(\mathbf{n}_i \cdot \mathbf{n}_j) \\
&+ (\mathbf{h}_j \cdot \mathbf{n}_i) - (\mathbf{h}_j \cdot \mathbf{n}_j)(\mathbf{n}_j \cdot \mathbf{n}_i)] \\
&- \frac{\beta}{4S}\sum_{ijl} \tilde{J}_{ij}\tilde{J}_{jl}[(\mathbf{n}_i \cdot \mathbf{n}_l) - (\mathbf{n}_i \cdot \mathbf{n}_j)(\mathbf{n}_j \cdot \mathbf{n}_l)] \\
&- \frac{\beta}{16S^2}\sum_{ij} \tilde{J}_{ij}^2 [1 - (\mathbf{n}_i \cdot \mathbf{n}_j)]^2. \qquad (20)
\end{aligned}$$

One can see that the effective classical Hamilton function corresponding to the Heisenberg quantum Hamiltonian, equation (11), 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 (15) generates terms of different topology in $\mathcal{H}^{(2)}$ (see Fig. 1)

$$\begin{aligned}
\mathcal{H}^{(2)} = &-\frac{\beta^2}{8S^2}\sum_{ijln} \tilde{J}_{ij}\tilde{J}_{jl}\tilde{J}_{ln}\Phi_{2\mathrm{a}}(\mathbf{n}_i, \mathbf{n}_j, \mathbf{n}_l, \mathbf{n}_n) \\
&- \frac{\beta^2}{12S^2}\sum_{ijln} \tilde{J}_{in}\tilde{J}_{jn}\tilde{J}_{ln}\Phi_{2\mathrm{b}}(\mathbf{n}_i, \mathbf{n}_j, \mathbf{n}_l, \mathbf{n}_n) \\
&- \frac{\beta^2}{24S^3}\sum_{ijl} \tilde{J}_{ij}^2\tilde{J}_{jl}\Phi_{2\mathrm{c}}(\mathbf{n}_i, \mathbf{n}_j, \mathbf{n}_l) \\
&- \frac{\beta^2}{48S^3}\sum_{ijl} \tilde{J}_{ij}\tilde{J}_{jl}\tilde{J}_{li}\Phi_{2\mathrm{d}}(\mathbf{n}_i, \mathbf{n}_j, \mathbf{n}_l) \\
&- \frac{\beta^2}{48S^4}\sum_{ij} \tilde{J}_{ij}^3\Phi_{2\mathrm{e}}(\mathbf{n}_i, \mathbf{n}_j). \qquad (21)
\end{aligned}$$

Here the 2a term is due to the first, second, and third terms of equation (15), the 2b term is due to the fourth term of equation (15), the 2c term is due to the fifth, sixth, and seventh terms of equation (15), the 2d term is due to the eighth term, and the 2e term is due to the ninth term. For the functions $\mathit{\Phi}$ in equation (21) we find

$$\begin{aligned}
\mathit{\Phi}_{2\mathrm{a}} &= \frac{16}{3}\Big[(\mathbf{n}_i\cdot\mathbf{n}_{j+})(\mathbf{n}_{j-}\cdot\mathbf{n}_{l+})(\mathbf{n}_{l-}\cdot\mathbf{n}_n)\\
&\quad+(\mathbf{n}_i\cdot\mathbf{n}_{j+})(\mathbf{n}_{j-}\cdot\mathbf{n}_{l-})(\mathbf{n}_{l+}\cdot\mathbf{n}_n)\\
&\quad+(\mathbf{n}_i\cdot\mathbf{n}_{j-})(\mathbf{n}_{j+}\cdot\mathbf{n}_{l+})(\mathbf{n}_{l-}\cdot\mathbf{n}_n)\Big]\\
&= (\mathbf{n}_i\cdot\mathbf{n}_n)-(\mathbf{n}_i\cdot\mathbf{n}_l)(\mathbf{n}_l\cdot\mathbf{n}_n)\\
&\quad-(\mathbf{n}_i\cdot\mathbf{n}_j)(\mathbf{n}_j\cdot\mathbf{n}_n)+(\mathbf{n}_i\cdot\mathbf{n}_j)(\mathbf{n}_j\cdot\mathbf{n}_l)(\mathbf{n}_l\cdot\mathbf{n}_n)\\
&\quad+\frac{1}{3}\big[(\mathbf{n}_i\cdot\mathbf{n}_l)(\mathbf{n}_j\cdot\mathbf{n}_n)-(\mathbf{n}_i\cdot\mathbf{n}_n)(\mathbf{n}_j\cdot\mathbf{n}_l)\big] \qquad (22)\\
\mathit{\Phi}_{2\mathrm{b}} &= 4(\mathbf{n}_i\cdot\mathbf{n}_{n+})(\mathbf{n}_j\cdot\mathbf{n}_n)(\mathbf{n}_l\cdot\mathbf{n}_{n-})\\
&= (\mathbf{n}_i\cdot\mathbf{n}_n)(\mathbf{n}_j\cdot\mathbf{n}_n)(\mathbf{n}_l\cdot\mathbf{n}_n)-\frac{1}{3}\Big[(\mathbf{n}_i\cdot\mathbf{n}_n)(\mathbf{n}_j\cdot\mathbf{n}_l)\\
&\quad+(\mathbf{n}_j\cdot\mathbf{n}_n)(\mathbf{n}_i\cdot\mathbf{n}_l)+(\mathbf{n}_l\cdot\mathbf{n}_n)(\mathbf{n}_i\cdot\mathbf{n}_j)\Big], \qquad (23)\\
\mathit{\Phi}_{2\mathrm{c}} &= -16\big[(\mathbf{n}_{i+}\cdot\mathbf{n}_{j+})(\mathbf{n}_{i-}\cdot\mathbf{n}_j)(\mathbf{n}_{j-}\cdot\mathbf{n}_l)\\
&\quad+(\mathbf{n}_{i-}\cdot\mathbf{n}_{j-})(\mathbf{n}_{i+}\cdot\mathbf{n}_j)(\mathbf{n}_{j+}\cdot\mathbf{n}_l)\\
&\quad+(\mathbf{n}_{i+}\cdot\mathbf{n}_{j+})(\mathbf{n}_{i-}\cdot\mathbf{n}_{j-})(\mathbf{n}_j\cdot\mathbf{n}_l)\big]\\
&= [1-(\mathbf{n}_i\cdot\mathbf{n}_j)]\big\{[3(\mathbf{n}_i\cdot\mathbf{n}_j)-1](\mathbf{n}_i\cdot\mathbf{n}_l)-2(\mathbf{n}_j\cdot\mathbf{n}_l)\big\}, \qquad (24)\\
\mathit{\Phi}_{2\mathrm{d}} &= 4^3(\mathbf{n}_{i+}\cdot\mathbf{n}_{j+})(\mathbf{n}_{j-}\cdot\mathbf{n}_{l+})(\mathbf{n}_{l-}\cdot\mathbf{n}_{i-})\\
&= (\mathbf{n}_i\cdot\mathbf{n}_j)^2+(\mathbf{n}_j\cdot\mathbf{n}_l)^2+(\mathbf{n}_l\cdot\mathbf{n}_i)^2\\
&\quad-(\mathbf{n}_i\cdot\mathbf{n}_j)(\mathbf{n}_j\cdot\mathbf{n}_l)(\mathbf{n}_l\cdot\mathbf{n}_i)\\
&\quad-\frac{1}{3}\Big[(\mathbf{n}_i\cdot\mathbf{n}_j)+(\mathbf{n}_j\cdot\mathbf{n}_l)+(\mathbf{n}_l\cdot\mathbf{n}_i)+(\mathbf{n}_i\cdot\mathbf{n}_j)(\mathbf{n}_j\cdot\mathbf{n}_l)\\
&\quad+(\mathbf{n}_j\cdot\mathbf{n}_l)(\mathbf{n}_l\cdot\mathbf{n}_i)+(\mathbf{n}_l\cdot\mathbf{n}_i)(\mathbf{n}_i\cdot\mathbf{n}_j)\Big], \qquad (25)
\end{aligned}$$

and

$$\begin{aligned}
\mathit{\Phi}_{2\mathrm{e}} &= 16(\mathbf{n}_{i+}\cdot\mathbf{n}_{j+})(\mathbf{n}_i\cdot\mathbf{n}_j)(\mathbf{n}_{i-}\cdot\mathbf{n}_{j-})\\
&= (\mathbf{n}_i\cdot\mathbf{n}_j)[1-(\mathbf{n}_i\cdot\mathbf{n}_j)]^2. \qquad (26)
\end{aligned}$$

The coefficient $\mathit{\Phi}_{2\mathrm{d}}$ can be rewritten in the form

$$\begin{aligned}
\mathit{\Phi}_{2\mathrm{d}} &= -[\mathbf{n}_i\mathbf{n}_j\mathbf{n}_l]^2\\
&\quad+[1-(\mathbf{n}_i\cdot\mathbf{n}_j)][1-(\mathbf{n}_j\cdot\mathbf{n}_l)(\mathbf{n}_l\cdot\mathbf{n}_i)], \qquad (27)
\end{aligned}$$

where $[\mathbf{abc}]\equiv\mathbf{a}\cdot(\mathbf{b}\times\mathbf{c})$ is the mixed product and an appropriate symmetrization is implied.

The field-dependent terms of order H^3, JH^2 and, J^2H in $\mathcal{H}^{(2)}$ can be calculated with the help of equation (16). The result has the form

$$\begin{aligned}
\mathcal{H}_h^{(2)} &= \frac{\beta^2}{12S^2}\sum_i(\mathbf{n}_i\cdot\mathbf{h}_i)[\mathbf{n}_i\times\mathbf{h}_i]^2\\
&\quad-\frac{\beta^2}{8S^2}\sum_{ij}\tilde{J}_{ij}\mathit{\Phi}_{2h^2}(\mathbf{n}_i,\mathbf{n}_j,\mathbf{h}_i,\mathbf{h}_j)\\
&\quad-\frac{\beta^2}{8S^2}\sum_{ijl}\tilde{J}_{ij}\tilde{J}_{jl}\mathit{\Phi}_{2h,\mathrm{a}}(\mathbf{n}_i,\mathbf{n}_j,\mathbf{n}_l,\mathbf{h}_i,\mathbf{h}_j,\mathbf{h}_l)\\
&\quad-\frac{\beta^2}{48S^3}\sum_{ij}\tilde{J}_{ij}^2\mathit{\Phi}_{2h,\mathrm{b}}(\mathbf{n}_i,\mathbf{n}_j,\mathbf{h}_i,\mathbf{h}_j), \qquad (28)
\end{aligned}$$

where

$$\begin{aligned}
\mathit{\Phi}_{2h^2} &= (\mathbf{n}_i\cdot\mathbf{n}_j)\big[(\mathbf{h}_i\cdot\mathbf{n}_i)^2+(\mathbf{h}_j\cdot\mathbf{n}_j)^2\big]\\
&\quad-\frac{2}{3}\big[(\mathbf{h}_i\cdot\mathbf{n}_i)(\mathbf{h}_i\cdot\mathbf{n}_j)+(\mathbf{h}_j\cdot\mathbf{n}_j)(\mathbf{h}_j\cdot\mathbf{n}_i)\big]\\
&\quad-\frac{1}{3}(\mathbf{n}_i\cdot\mathbf{n}_j)[h_i^2+h_j^2]\\
&\quad+(\mathbf{h}_i\cdot\mathbf{h}_j)-(\mathbf{h}_i\cdot\mathbf{n}_i)(\mathbf{n}_i\cdot\mathbf{h}_j)-(\mathbf{h}_i\cdot\mathbf{n}_j)(\mathbf{n}_j\cdot\mathbf{h}_j)\\
&\quad+(\mathbf{h}_i\cdot\mathbf{n}_i)(\mathbf{n}_i\cdot\mathbf{n}_j)(\mathbf{n}_j\cdot\mathbf{h}_j)\\
&\quad+\frac{1}{3}\big[(\mathbf{h}_i\cdot\mathbf{n}_j)(\mathbf{h}_j\cdot\mathbf{n}_i)-(\mathbf{h}_i\cdot\mathbf{h}_j)(\mathbf{n}_i\cdot\mathbf{n}_j)\big], \qquad (29)\\
\mathit{\Phi}_{2h,\mathrm{a}} &= (\mathbf{h}_i\cdot\mathbf{n}_l)-(\mathbf{h}_i\cdot\mathbf{n}_j)(\mathbf{n}_j\cdot\mathbf{n}_l)\\
&\quad+(\mathbf{h}_l\cdot\mathbf{n}_i)-(\mathbf{h}_l\cdot\mathbf{n}_j)(\mathbf{n}_j\cdot\mathbf{n}_i)\\
&\quad-[(\mathbf{h}_i\cdot\mathbf{n}_i)+(\mathbf{h}_l\cdot\mathbf{n}_l)][(\mathbf{n}_i\cdot\mathbf{n}_l)-(\mathbf{n}_i\cdot\mathbf{n}_j)(\mathbf{n}_j\cdot\mathbf{n}_l)]\\
&\quad+\frac{1}{3}\Big[(\mathbf{n}_i\cdot\mathbf{n}_l)\big[(\mathbf{h}_i\cdot\mathbf{n}_j)+(\mathbf{h}_l\cdot\mathbf{n}_j)\big]\\
&\quad-(\mathbf{h}_i\cdot\mathbf{n}_l)(\mathbf{n}_i\cdot\mathbf{n}_j)-(\mathbf{h}_l\cdot\mathbf{n}_i)(\mathbf{n}_l\cdot\mathbf{n}_j)\Big]\\
&\quad+2(\mathbf{n}_i\cdot\mathbf{n}_j)(\mathbf{h}_j\cdot\mathbf{n}_j)(\mathbf{n}_j\cdot\mathbf{n}_l)-\frac{2}{3}\Big[(\mathbf{n}_i\cdot\mathbf{n}_l)(\mathbf{h}_j\cdot\mathbf{n}_j)\\
&\quad+(\mathbf{n}_j\cdot\mathbf{n}_l)(\mathbf{h}_j\cdot\mathbf{n}_i)+(\mathbf{n}_j\cdot\mathbf{n}_i)(\mathbf{h}_j\cdot\mathbf{n}_l)\Big], \qquad (30)
\end{aligned}$$

and

$$\begin{aligned}
\mathit{\Phi}_{2h,\mathrm{b}} &= -[1-(\mathbf{n}_i\cdot\mathbf{n}_j)]\big\{2[(\mathbf{h}_i\cdot\mathbf{n}_j)+(\mathbf{h}_j\cdot\mathbf{n}_i)]\\
&\quad+[1-3(\mathbf{n}_i\cdot\mathbf{n}_j)][(\mathbf{h}_i\cdot\mathbf{n}_i)+(\mathbf{h}_j\cdot\mathbf{n}_j)]\big\}. \qquad (31)
\end{aligned}$$

For a homogeneous field, $\mathbf{h}_i=\mathbf{h}$, the last expression becomes

$$\mathit{\Phi}_{2h,\mathrm{b}} = -3[\mathbf{h}\cdot(\mathbf{n}_i+\mathbf{n}_j)][1-(\mathbf{n}_i\cdot\mathbf{n}_j)]^2. \qquad (32)$$

As was said after equation (10), cumulant expansion is, in general, an expansion in β/S. In particular, in zero field in the nth order of the cumulant expansion, terms of order $\tilde{J}(\beta\tilde{J}/S)^{n-1}$ appear. On the other hand, there are also terms carrying additional powers of $1/S$, such as the last term of equation (20) and the last three terms of equation (21). This feature results, formally, from reexpression of cumulants of composite operators by ordinary cumulants, equations (14, 15), which are specific to many-spin systems. For $S \gg 1$, the terms containing powers of $\beta\tilde{J}/S$ without additional factors $1/S$ dominate over the other terms in the whole range of temperatures. Those terms are given by "tree" diagrams, such as 0, 1a, 2a, and 2b in Figure 1. These diagrams are maximally branched and they do not contain loops or parallel interaction lines. Non-tree diagrams can be obtained from the tree diagrams by joining two solid circles into one. This leads each time to appearance of a spin cumulant of a higher order and thus to an additional factor of $1/S$ (see Eq. (9)). Minimally branched diagrams are those consisting of several parallel wavy lines, such as diagrams 1b and 2e in Figure 1. These diagrams result in terms of order $\tilde{J}(\beta\tilde{J}/S^2)^{n-1}$ in $\mathcal{H}$.

5 Physical quantities

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

$$U = -\partial \ln \mathcal{Z}/\partial\beta = \langle \mathcal{H}^* \rangle, \tag{33}$$

where $\langle \dots \rangle$ denotes a classical thermal average and

$$\mathcal{H}^* = \partial(\beta\mathcal{H})/\partial\beta = \mathcal{H}^{(0)} + 2\mathcal{H}^{(1)} + 3\mathcal{H}^{(2)} + \dots \tag{34}$$

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

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

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

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

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

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

One notices that in order to calculate a correlation function, it is insufficient to perform a classical thermal average of $n_{i\alpha}n_{j\beta}$ or even of $n^*_{i\alpha}n^*_{j\beta}$. The last term of equation (37) makes a contribution to the third order in the cumulant expansion due to the terms of type JH^2 (see Eq. (28)).

6 Application to the spin chain

The isotropic spin chain in zero magnetic field is a simple solvable model in the classical limit [9]. 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

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

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

$$\tilde{\mathcal{Z}}_0 = \left(\frac{2S+1}{4\pi}\right)^N \mathcal{Z}_0, \tag{39}$$

and $\mathcal{Z}_0$ is the partition function of the classical system. For the open spin chain, the latter is given by $\mathcal{Z}_0 = 4\pi(4\pi\sinh(\xi)/\xi)^{N-1}$ with $\xi \equiv \beta\tilde{J}$. To order $1/S^2$, one should use for the linear $\mathcal{H}^{(1)}$ term in equation (38) the third and fourth terms of equation (20), for the quadratic $\mathcal{H}^{(1)}$ term the third term of equation (20), and for the linear $\mathcal{H}^{(2)}$ term the first and second terms of equation (21). After performing thermodynamic averages one obtains

$$\frac{\ln\mathcal{Z}}{N} \cong \ln(2S+1) + \ln\left(\frac{\sinh\xi}{\xi}\right) + \frac{\xi B}{S} + \frac{5\xi^2 - 7\xi^2B^2 \mp \xi^2B - 9\xi B}{12S^2} + O\left(\frac{1}{S^3}\right), \tag{40}$$

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, equation (20) of reference [3], where the factor 7 is replaced by 6. For the energy per spin $U = -\partial\ln\mathcal{Z}/\partial(\beta N)$ one obtains

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

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

$$U = -\frac{\tilde{J}_0}{2} - \frac{\tilde{J}_0}{2S}\int\frac{\mathrm{d}\mathbf{q}}{(2\pi)^3}(1 - \tilde{\varepsilon}_{\mathbf{q}}) + T\int\frac{\mathrm{d}\mathbf{q}}{(2\pi)^3}\frac{\tilde{\beta}\tilde{\varepsilon}_{\mathbf{q}}}{\exp(\tilde{\beta}\tilde{\varepsilon}_{\mathbf{q}}) - 1}$$

$$\tilde{\beta} \equiv \beta\tilde{J}_0/S, \quad \tilde{\varepsilon}_{\mathbf{q}} \equiv (S/\tilde{J}_0)\varepsilon_{\mathbf{q}} = \begin{cases} 1 - \lambda_{\mathbf{q}}, & \mathrm{F} \\ \sqrt{1 - \lambda_{\mathbf{q}}^2}, & \mathrm{AF} \end{cases}. \tag{42}$$

Here $\tilde{J}_0$ is the zeroth Fourier component of $\tilde{J}_{ij}$ and $\lambda_{\mathbf{q}} \equiv \tilde{J}_{\mathbf{q}}/\tilde{J}_0$. 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

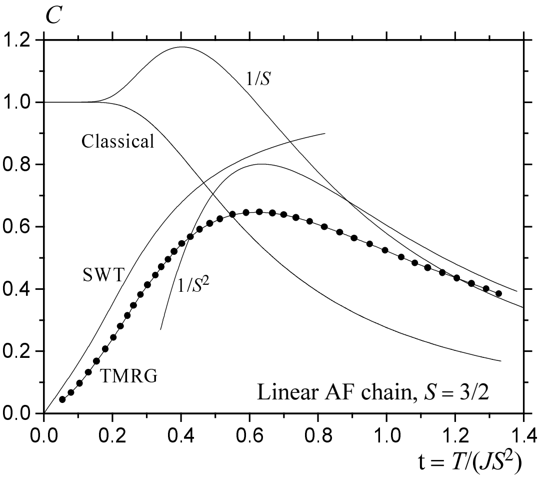

Fig. 3. Heat capacity of the antiferromagnetic Heisenberg $S = 3/2$ chain. Theoretical results to zeroth, first and second order of the cumulant expansion, as well as of linear spin-wave theory (SWT). They are compared with the numerical TMRG calculation of reference [10].

the temperature-dependent magnon contribution. Strictly speaking, linear spin-wave theory is only applicable to more than two dimensions, although, equation (42) remains well-defined in lower dimensions. The Haldane gap in the magnon spectrum for integer S, which is not taken into account in equation (42), behaves as $\exp(-S)$ and becomes negligible for large spins. Whereas $\tilde{\varepsilon}_{\mathbf{q}}$ 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 that following from the cumulant expansion in the limit $\xi \equiv \tilde{J}/T \gg 1$. In particular, for a ferromagnetic chain in the temperature interval $\tilde{J}/S \ll T \ll \tilde{J}$ equation (41) yields

$$\frac{U}{\tilde{J}} \cong -1 - \frac{1}{S} + \frac{1}{\xi} + \frac{\xi}{2S^2} - \frac{1}{2S^2}, \qquad (43)$$

whereas expanding equation (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 Figure 3 we compare the heat capacity $C = \mathrm{d}U/\mathrm{d}T$ of a Heisenberg antiferromagnetic chain with $S = 3/2$ calculated to different orders of the cumulant expansion (see Eq. (41)) and following from the linear spin-wave theory (see Eq. (42)) with the numerical result of reference [10] where the transfer-matrix renormalization group (TMRG) was used. Although a spin value of 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 the calculation of the next terms of the effective classical Hamiltonian, which can be done with the help of the diagram technique developed in Section 3. Also on the low-temperature side the accuracy of the SWT result can be improved by taking into account in equation (42), the $1/S$ corrections to the magnon spectrum.

7 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 $\hat{H}$, equation (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 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 reference [8]. 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 reference [8] 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. [11]), which, however, should be generalized to non-Heisenberg Hamiltonians. 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 [12–14]. 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,

$$\sum_{\alpha=x,y} (\mathbf{a}\cdot\mathbf{n}_\alpha)(\mathbf{n}_\alpha\cdot\mathbf{b}) = (\mathbf{a}\cdot\mathbf{b}) - (\mathbf{a}\cdot\mathbf{n})(\mathbf{n}\cdot\mathbf{b}), \qquad (A.1)$$

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

$$(\mathbf{a}\cdot\mathbf{n}_x)(\mathbf{n}_y\cdot\mathbf{b}) - (\mathbf{a}\cdot\mathbf{n}_y)(\mathbf{n}_x\cdot\mathbf{b}) = ([\mathbf{n}_x\times\mathbf{n}_y]\cdot[\mathbf{a}\times\mathbf{b}]) = (\mathbf{n}\cdot[\mathbf{a}\times\mathbf{b}]). \quad (A.2)$$

Combining these two formulas one obtains the relation

$$4(\mathbf{a}\cdot\mathbf{n}_\pm)(\mathbf{n}_\mp\cdot\mathbf{b}) = (\mathbf{a}\cdot\mathbf{b}) - (\mathbf{a}\cdot\mathbf{n})(\mathbf{n}\cdot\mathbf{b}) \pm \mathrm{i}(\mathbf{n}\cdot[\mathbf{a}\times\mathbf{b}]), \quad (A.3)$$

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

$$(\mathbf{n}_+\cdot\mathbf{n}_-) = \frac{1}{4}(n_x^2+n_y^2) = \frac{1}{2}$$

$$[\mathbf{n}_+\times\mathbf{n}_-] = \frac{\mathrm{i}}{2}\mathbf{n}, \quad [\mathbf{n}_\pm\times\mathbf{n}] = \mp\mathrm{i}\mathbf{n}_\pm. \quad (A.4)$$

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