Heat capacity of anisotropic Heisenberg antiferromagnet within the spin Hartree-Fock approach in quasi-1D Regime

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Abstract We study the anisotropic quantum Heisenberg antiferromagnet for spin-1/2 that interpolates smoothly between the one-dimensional (1D) and the two-dimensional (2D) limits. Using the spin Hartree-Fock approach we construct a quantitative theory of heat capacity in the quasi-1D regime with a finite coupling between spin chains. This theory reproduces closely the exact result of Bethe Ansatz in the 1D limit and does not produces any spurious phase transitions for any anisotropy in the quasi-1D regime at finite temperatures in agreement with the Mermin-Wagner theorem. We study the static spin-spin correlation function in order to analyse the interplay of lattice geometry and anisotropy in these systems. We compare the square and triangular lattice. For the latter we find that there is a quantum transition point at an intermediate anisotropy of \( \sim 0.6 \). This quantum phase transition establishes that the quasi-1D regime extends up to a particular point in this geometry. For the square lattice the change from the 1D to 2D occurs smoothly as a function of anisotropy, i.e. it is of the crossover type. Comparing the newly developed theory to the available experimental data on the heat capacity of \( \text{Cs}_2\text{CuBr}_4 \) and \( \text{Cs}_2\text{CuCl}_4 \) we extract the microscopic constants of the exchange interaction that previously could only be measured using inelastic neutron scattering in high magnetic fields.

1 Introduction

The low-dimensional quantum antiferromagnets have attracted a significant degree of attention in the last two decades due to synthesis of an increasing number of anisotropic magnetic insulators with dimensionalities between one and two, such as \( \text{CsNiCl}_3 \), \( \text{Cs}_2\text{CuCl}_4 \), \( \text{CuBr}_4 \), and \( \text{Cs}_2\text{CuBr}_4 \). The dimensionality of these materials is larger than strictly one where the exact Bethe ansatz methods are available [5] but is below three where the classical antiferromagnetic order was rigorously proven [6,7], requiring an accurate description of the quantum effects in the anisotropic 2D antiferromagnets. Popular methods to deal with this problem are the Takahashi’s modified spin-wave theory [8] and the mean-field theory based on the Schwinger-boson representation of spin-waves by Arovas and Auerbach [9]. These approaches are quite successful particularly for ferromagnets, for which they produce the correct subleading terms of the free energy in 1D [8] that were obtained by means of the exact thermodynamic Bethe ansatz [10,11]. However, their predictions are not so good for \( S = 1/2 \) antiferromagnets, for which they predict a spurious phase transition at finite temperatures in 1D and 2D, which is explicitly forbidden in these dimensions by the Mermin-Wagner theorem [12]. Thus, description of thermodynamic quantities, like the heat capacity, in these systems in the quasi-1D regime, where a finite interaction between the spin chains makes the exact Bethe ansatz already inapplicable, at low to intermediate temperatures requires a different approach. Other proposed methods include RVB theory [13,14], Wigner-Jordan fermions [15], interpolation based on the high-temperature expansions [16], and spin-Hartree Fock approach [17].

In this paper, we use the recently proposed spin Hartree-Fock approach [17] in order to construct a theory for the heat capacity of the anisotropic Heisenberg model for spin-1/2 on the square and triangular lattices in the quasi-1D regime with two different exchange energies \( J \) and \( J' \) along the direction of the strongest coupling and in the other direction respectively. Within this theory we evaluate the temperature dependence of heat capacity and establish applicability of the quasi-1D regime for different degrees of anisotropy, using the heat capacity itself at low temperatures and the next-neighbour correlation functions. We assess the quality of this theory quantitatively by fitting the obtained temperature dependence of heat capacity to the available experimental data for \( \text{Cs}_2\text{CuCl}_4 \) and \( \text{Cs}_2\text{CuBr}_4 \). The result for the microscopic exchange energies in the Heisenberg model obtained using these fits matches well the well-known values obtained in neutron scattering experiment available for this materials [18,19], confirming the validity of our approach.

The rest of the paper is organised as follows. In Section 2, we describe the anisotropic Heisenberg model on the square and on the triangular lattice. Section 3 contains application of the spin Hartree-Fock approach to the model on the triangular lattice and derivation of the self-
consistency equations for the corresponding mean-field parameters. In Section 4, we derive the explicit expression for the free energy within the spin Hartree-Fock approach and analyse the heat capacity as a function of temperature and anisotropy parameter for the triangular lattice in detail. In Section 5 we evaluate the static spin-spin correlation function for the triangular lattice and highlight the differences with the triangular lattice. In Section 6, we fit the heat capacity experimentally measured for a pair of magnetic insulators with the theory developed in Sections 3 and 4.

2 Heisenberg Hamiltonian on anisotropic 2D lattices

The Heisenberg model for spin-1/2 on the anisotropic 2D lattice in the absence of an external magnetic field reads

$$H = \frac{J}{2} \sum_{\mathbf{r}, \delta_1} \mathbf{S}_\mathbf{r} \cdot \mathbf{S}_{\mathbf{r} + \delta_1} + \frac{J'}{2} \sum_{\mathbf{r}, \delta_2} \mathbf{S}_\mathbf{r} \cdot \mathbf{S}_{\mathbf{r} + \delta_2},$$

where $J$ and $J'$ are two exchange energies that are different along a pair of primitive vectors of a 2D lattice, $S^x_\mathbf{r}, S^y_\mathbf{r}, S^z_\mathbf{r}$ are the spin-1/2 operators at site $\mathbf{r}$, the sum over $\mathbf{r}$ runs over a 2D lattice consisting of $N = L^2$ spins, and the sums over $\delta_1$ and $\delta_2$ run over the nearest-neighbours connected by the exchange interactions $J$ and $J'$ respectively. We impose the periodic boundary conditions, $\mathbf{S}_{\mathbf{r} + \delta_1(e_2)} L = \mathbf{S}_\mathbf{r}$, and restrict ourselves to the antiferromagnetic exchange energies, $J > 0$ and $J' > 0$.

In order to compare the effect of different lattice geometries we consider two particular 2D Bravais lattices in this work. One is the square lattice as the simplest. The primitive vectors in the units of lattice parameter are $\mathbf{e}_1 = x$ and $\mathbf{e}_2 = y$, see Fig. 1(a). There are two nearest-neighbours along the principle direction with the exchange coupling constant $J$ and two nearest-neighbours in the perpendicular direction with $J'$. Apart from the square lattice we will also consider the triangular lattice in this work. The primitive vectors in the units of lattice parameter are $\mathbf{e}_1 = x$ and $\mathbf{e}_2 = x/2 + \sqrt{3}y/2$, see Fig. 1(b). There are two nearest-neighbours along the principle direction with the exchange coupling constant $J$ and the other four nearest-neighbours out of the total of six on this lattice with $J'$. The precise phase diagram for arbitrary $J'/J$ is still not fully established, see a review in [19]. Below, we consider the triangular lattice in detail motivated by a pair of magnetic insulators, Cs$_2$CuCl$_4$ and Cs$_2$CuBr$_4$. The analysis of the square lattice is very similar and is concisely presented in Section 6.

3 Spin Hartree-Fock approach

In order to analyse the model in Eq. 1 we employ the spin Hartree-Fock approach developed in Ref. [17]. Using this method in the present Section we derive the self-consistency equations for the mean-field parameters in the anisotropic case for the triangular lattice. Solutions of these equations as a function of temperature and the degree of anisotropy are used later to analyse the thermodynamics and correlation functions in Sections 5 and 6.
The number of the independent spin components in Eq. (1) can be reduced since the spin-1/2 operators on the same site satisfy the following identity,
\[ S_r^+ S_r^- = S_r^+ S_r^- - 1/2, \] (2)
in addition to the angular momentum commutation relations \[ [S_{\alpha}^+, S_{\beta}^-] = i\varepsilon_{\alpha\beta\gamma} S_r^\gamma, \] where \( \varepsilon_{\alpha\beta\gamma} \) is the Levi-Civita symbol of third rank. Substituting Eq. (2) in the \( z \)-component of the scalar products in Eq. (1) and performing a Fourier transform of the whole Hamiltonian by substituting \( S_r^\pm = N^{-1/2} \sum_k S_k^\pm e^{\pm i k \cdot r} \) we obtain a sum of a quadratic and a quartic form in the spin operators in the momentum space,
\[ H = \sum_k \left[ J (\varepsilon_1 (k) - 1) + 2J' (\varepsilon_2 (k) - 1) \right] S_k^+ S_k^- + \frac{1}{N} \sum_{k_1,k_2,k_3,k_4} \left[ J \varepsilon_1 (k_3 - k_4) + 2J' \varepsilon_2 (k_3 - k_4) \right] \times \delta_{k_1+k_3,k_2+k_4} S_{k_1}^+ S_{k_2}^+ S_{k_3}^- S_{k_4}^- \] (3)
where \( \varepsilon_1 (k) = \cos k_x \) and \( \varepsilon_2 (k) = \cos (k_x/2) \cos (\sqrt{3} k_y/2) \) are the dispersions along the primitive vectors \( e_1 \) and \( e_2 \). Here we set the lattice and the Planck constants to unity, \( a = 1 \) and \( \hbar = 1 \) respectively.

The structure of the model in Eq. (3) resembles that of a model of interacting particles but for spin operators. The first (quadratic term) could be interpreted as a kinetic energy and the second (quartic term) as a two-body interaction between magnetic excitations. Thus, we analyse the model in Eq. (3) using a spin variety of the Hartree-Fock approximation. Following Ref. [17], we make an assumption that the many-body eigenstates can be approximated by product states of the single spin magnetic excitations when the number of spins \( N \) is very large in the thermodynamic limit. Taking into account finite temperature, such an approximation corresponds to a product density matrix, \( \rho = \prod_k m_k |k \rangle |1 - m_k \rangle \), where \( |k \rangle \) is a state of a single magnetic excitation with momentum \( k \), the scalar parameters \( m_k \) are free parameters that will be defined later, and the normalisation of the density matrix is chosen such that \( \text{Tr} \rho = 1 \).

The free parameters \( m_k \) can obtained in the usual to mean-field way by minimising the free energy,
\[ F = E - TS, \] (4)
at an arbitrary temperature \( T \). The expectation value of Eq. (3) with respect to \( \rho \) gives the expression for the energy of the system \( E = \text{Tr} \{ H \rho \} \) as a function of parameters \( m_k \),
\[ E = \sum_k \left[ J (\varepsilon_1 (k) - 1) + 2J' (\varepsilon_2 (k) - 1) \right] m_k \]
\[ -\frac{1}{N} \sum_{k_1,k_2} \left( J \varepsilon_1 (k_1 - k_2) + 2J' \varepsilon_2 (k_1 - k_2) \right) m_{k_1} m_{k_2}, \] (5)

The von Neumann entropy, \( S = -k_B \text{Tr} [\rho \ln \rho] \), can be expressed in terms of the parameters \( m_k \) as follows,
\[ S = -k_B \sum_k [m_k \ln m_k + (1 - m_k) \ln (1 - m_k)], \] (6)
where \( k_B \) is the Boltzmann constant. Then the solution of the minimisation problem, \( \partial F / \partial m_k = 0 \), gives the self-consistency equations for each \( m_k \) in the form of a large set of \( N \) nonlinear equations,
\[ m_k = \frac{1}{\exp \left( \frac{E(k)}{k_B T} \right) + 1}, \] (7)
where the energy in the exponential for every \( m_k \) depends on all the other mean-field parameters as
\[ E (k) = J (\varepsilon_1 (k) - 1) + 2J' (\varepsilon_2 (k) - 1) - \frac{2}{N} \sum_{k'} [J \varepsilon_1 (k - k') + 2J' \varepsilon_2 (k - k')] m_{k'}. \] (8)

Since \( m_k \) in the set of equations in Eq. (7) enter only under a sum, the number of equations can be reduced to just a few. Introduction of the following three extensive variables,
\[ s = \frac{1}{N} \sum_k m_k - \frac{1}{2}, \] (9)
\[ u_\alpha = -\frac{1}{N} \sum_k \varepsilon_\alpha (k) m_k + \frac{1}{2}, \] (10)
where \( \alpha = 1, 2 \) corresponds to the primitive vectors \( e_1 \) and \( e_2 \), reduces Eq. (7) to a set of only three independent equations,

\[
s = \int \frac{d^2 k}{V} \exp \left[ \frac{1}{2(2J + 4J')} s + 2Ju_{1\varepsilon_1}(k) + 4J'u_{2\varepsilon_2}(k) / (k_BT) \right] + \frac{1}{2},
\]

\[
u_{\alpha} = \frac{1}{2} - \int \frac{d^2 k}{V} \exp \left[ \frac{1}{2(2J + 4J')} s + 2Ju_{1\varepsilon_1}(k) + 4J'u_{2\varepsilon_2}(k) / (k_BT) \right] + 1,
\]

where the thermodynamic limit was taken as \( \sum k/N \to \int d^2 k/V \), the momentum integral runs over the first Brillouin zone, and \( V = 8\pi^2/\sqrt{3} \) is the volume of the primitive cell in the reciprocal space of the triangular lattice. A renormalised single-particle like dispersion in the exponent of the equations in Eq. (11) and (12) reproduces qualitatively the Bethe ansatz result [24] and gives the leading contribution to observables in the thermodynamic limit at high energy [25,26,27].

The system of nonlinear equations in Eq. (11) and (12) can be solved numerically. For \( J' = 0 \) the equation for \( u_2 \) drops out (the value \( u_2 \equiv 1/2 \) becomes independent of all other parameters) since the system becomes one-dimensional. The two remaining equations for \( s \) and \( u_1 \) can be solved explicitly at zero temperature, \( T = 0 \). The integrands are proportional to the Heaviside step function,

\[
\lim_{T \to 0} \frac{1}{\varepsilon^2/(k_BT) + 1} = \Theta(-x),
\]

and the integrals can be evaluated explicitly. Solution of the resulting linear equations immediately gives only one solution \( s = 0 \) and \( u_1 = 1/2 + 1/\pi \). Starting from this point Eqs. (11) and (12) can be continuously deformed in a smooth way to arbitrary values of \( J'/J \) and \( T \) giving \( s \) and \( u_1 \) as functions of \( J'/J \) and \( T \). For all values of \( J'/J \) and \( T \) this procedure gives \( s = 0 \), i.e., the net magnetisation of the antiferromagnetic system is always zero. The thermodynamic observables and the static correlation functions can be expressed in terms of the two functions \( u_{1,2}(J'/J, T) \). We will analyse some representative examples of them in the next two Sections.

## 4 Thermodynamics

Now we use the mean-field equations obtained in the previous Section to analyse the thermodynamic properties of the model in Eq. (1). First, we derive the general expression for the free energy in terms of the mean-field parameters \( s \) and \( u_1 \) that can be used to evaluate all thermodynamic quantities. Then, we use a particular thermodynamic observable, the specific heat \( C \), to study in detail the dependencies on the anisotropy parameter \( J'/J \) and temperature \( T \).

The free energy in Eq. (9) is expressed in terms of \( E \) and \( S \) that, in turn, can be expressed through the three mean-field parameters \( s \) and \( u_1 \) and temperature \( T \). Using the definitions in Eqs. (9) and (10), the energy of the system \( E \) in Eq. (5) can be written as

\[
E = N \left[ (J + J') s^2 - Ju_1^2 - 2J'u_2^2 \right].
\]

The expression for the entropy is more complicated. The energy in the exponential in Eq. (3) can be expressed, analogously to Eq. (13), using the definitions in Eqs. (9) and (10) in a simple way as

\[
\mathcal{E}(k) = (2J + 4J') s + 2Ju_{1\varepsilon_1}(k) + 4J'u_{2\varepsilon_2}(k).
\]

The von Neumann entropy in Eq. (9) can then be written in terms of this \( \mathcal{E}(k) \) as

\[
S = Nk_BT \int \frac{d^2 k}{V} \ln \left[ \frac{\mathcal{E}(k)}{2k_BT} \right] + Nk_BT \int \frac{d^2 k}{V} \ln \left[ \cosh \left( \frac{\mathcal{E}(k)}{2k_BT} \right) \right].
\]

Substitution of Eqs. (13) and (16) into Eq. (18) gives the expression for the free energy in terms of only the mean-field parameters, the microscopic parameters of the original model in Eq. (1), and temperature. All the thermodynamic observables can be derived from it using the general thermodynamic identities and derivatives with respect to microscopic parameters and temperature. However, the resulting expressions obtained using this generic method are not very compact and are rather complicated due to the integrals over momentum in the expression for entropy in Eq. (16).

Here we focus on the specific thermodynamic observable—the heat capacity \( C \). It can be expressed through \( s \) and \( u_1 \) in a simpler way using its original definition,

\[
C = \frac{\partial E}{\partial T},
\]

instead of taking a second derivative of the free energy obtained above,

\[
C = -T \frac{\partial^2 F}{\partial T^2}.
\]

Substitution of the energy \( E \) in Eq. (14) into Eq. (17) gives the following expression for the heat capacity per spin,

\[
C/N = (2J + 4J') \frac{\partial s}{\partial T} - 2Ju_1 \frac{\partial u_1}{\partial T} - 4J'u_2 \frac{\partial u_2}{\partial T}.
\]

This expression, together with the numerical solutions of Eqs. (11) and (12) for various values of \( T \) and \( J'/J \) and where the derivative with respect to temperature is evaluated numerically, is used for obtaining the plots in Fig. 2.
The temperature dependence of $C$ for different values of the anisotropy $J'/J$ ranging from $J'/J = 0$ (the 1D limit) to $J'/J = 1$ (the 2D limit) is plotted in Fig. 2(a). In the 1D limit a 2D magnet splits into a set of independent 1D magnets that are completely isolated from each other. For each 1D magnet the 1D Heisenberg model for spin-1/2 can be diagonalised exactly using the Bethe ansatz approach and the heat capacity can also be evaluated using this diagonalisation procedure without any approximation. The exact result reproduces quite well the $J'/J = 0$ (the 1D limit) curve in Fig. 2(a), especially at low temperatures up to $T/J \sim 0.35$. In this region quantitative difference between the exact result and the result of the spin Hartee-Fock approach is negligible. In the region from $T/J \approx 0.35$ to $T/J \approx 1$ deviations are still appreciable, of the order of 15%. The accuracy in this region can be improved by taking into account systematically the correlation function of order higher than two, e.g. using the recently proposed spin-FRG approach.

For all values of the anisotropy there is no sign of any finite temperature phase transition, i.e. there are no signs of a singularity or of a kink. This behaviour is in full accord with the Mermin-Wagner theorem that explicitly forbids a long-range order parameter for the spin-1/2 Heisenberg magnets at finite temperatures in two and lower dimensions. In the low temperature limit, $T \to 0$, the heat capacity remains a linear function of $T$, $C \approx \gamma T$, when a small but finite $J'$ is introduced. The dependence of $\gamma$ on the anisotropy parameter $J'/J$ is plotted in Fig. 2(b). At an intermediate anisotropy of $J' \approx 0.6198J$ the linear coefficient at low temperatures has a cusp signalling a quantum phase transition for the triangular lattice, which establishes the validity of the quasi-1D regime. It will be analysed in more detail using the static correlation function that we evaluate in the next section.

The conventional spin-wave theory gives a quadratic dependence in $T$ dependence in the 2D regime for $J' > 0.6198J$. However, we are concentrated on the quasi-1D regime $J < 0.6198J$ in this work. This value of $J'_{\text{crit}} = 0.6198J$ is close to values of $J'_{\text{crit}} \approx 0.6J$ obtained by other methods.

5 Correlation functions

In this Section we analyse the static spin-spin correlation function of the model in Eq. (1). We derive the general expression for the static finite-range spin-spin correlation function within the Hartree-Fock approach. Then, we study in detail its next-neighbour part along the principal axis of the anisotropic triangular lattice as a function of the anisotropy parameter $J'/J$.

The static spin-spin correlation function can be evaluated as an expectation value of the spin operator $S_0 \cdot S_r$ with respect to the density matrix in Eq. (7). The result depends on many mean-field parameters $m_k$. Using the definitions in Eqs. (11) and (12), the expression can be reduced to only a function of the three extensive parameters $s$ and $u$.

\[
\langle S_0 \cdot S_r \rangle = s^2 + I(r) [1 - I(r)], \tag{20}
\]

where the function that depends on the coordinate is

\[
I(r) = \int \frac{d^2 k}{V} \frac{\cos (k \cdot r)}{\exp \left[ \left< (2J + 4J') s + 2J u_{1e_1}(k) + 4J' u_2 e_2(k) \right>/ (k_B T) \right] + 1}. \tag{21}
\]
Here we consider the model in Eq. (1) for the anisotropic square lattice in Fig. 1(a). We quickly sketch the application of the spin Hartree-Fock approach in this case that is almost identical to the triangular lattice considered in Sections 3-5 and quote the corresponding results for the heat capacity and for the next-neighbour part of the static spin-spin correlation function. Then, we conclude by analysing numerically these two observables and highlight the differences in them between the square and the triangular lattices.

In this section we consider the anisotropic square lattice in Fig. 1(a). The two primitive vectors in the real space are now orthogonal, $e_1 = x$ and $e_2 = y$. The Fourier transform of the model in Eq. (1) on this lattice gives

$$H = \sum_k [J(\varepsilon_1(k) - 1) + J'(\varepsilon_2(k) - 1)] S_k^+ S_k^- + \frac{1}{N} \sum_{k_1, k_2, k_3, k_4} [J\varepsilon_1(k_3 - k_1) + J'\varepsilon_2(k_3 - k_4)] 
\times \delta_{k_1 + k_2 + k_3 - k_4} S_{k_1}^+ S_{k_2}^- S_{k_3}^+ S_{k_4}^-.$$  

(23)

This value is also close the exact diagonalisation result $(S_0 \cdot S_{e_1}) = -0.182 \ldots$ obtained in [23]. In between there is a quantum transition point identified as a finite jump in the correlation function at $J'_\text{crit} \approx 0.6198 J$, which is within the range of $J'_\text{crit}$ from 0.60 J to 0.65 J obtained by a quantum Monte Carlo approach [21] and by using the RVB ansatz [22][23]. Below this point ($J < J'_\text{crit}$) there is a quasi-1D order close to the strictly 1D Bethe ansatz solution with weak correlations between the chains.

6 Square lattice

Note that the coordinate $r$ above labels a discrete set of nodes on the triangular lattice, i.e. $r = e_1 i + e_2 j$ where $i$ and $j$ integer numbers. At $T = 0$ the expression in Eqs. (20) and (21) gives correlations that vanish as a power-law and at finite $T$ it gives produces an exponential behaviour at long distances, see details and discussion in Ref. [17].

The signatures of the classical order, however, still survive in the next-neighbour part of the correlation function. In this case the integral in Eq. (21) simplifies even further by use of the definitions in Eqs. (9) and (10) giving $I(e_1) = 1/2 - u_1$ and $I(e_2) = 1/2 - u_2$. Then, the next-neighbour correlation function along the principle direction of the triangular lattice in Fig. 1(b) obtained by the substitution of these expressions into Eq. (20) reads as

$$\langle S_0 \cdot S_{e_1(e_2)} \rangle = s^2 - u_1^2 + \frac{1}{4}. \quad (22)$$

The above expression with the solution of Eqs. (11) and (12) as a function of the anisotropy parameter $J'/J$ at zero temperature $T = 0$ is plotted in Fig. 3. In the 1D limit ($J'/J = 0$) the next-neighbour correlation function along the chains (blue line) is $(S_0 \cdot S_{e_1}) = -0.4196 \ldots$ that is close to the Bethe ansatz result $(S_0 \cdot S_{e_1}) = -0.4431 \ldots$ marked by the thick black line.

In the $J'/J = 1$ limit the next-neighbour correlation function in all direction is $(S_0 \cdot S_{e_1}) = (S_0 \cdot S_{e_2}) = -0.2017 \ldots$ that is reminiscent of the classical 120-degree order $(S_0 \cdot S_{e_1}) = (S_0 \cdot S_{e_2}) = -0.125$ but still shows a relatively large enhancement due to quantum fluctuations.

Figure 4. Specific heat $C$ as a function of $T$ for the anisotropic square lattice for a set of different values of the anisotropy parameter $J'/J = 0$ (blue line), 0.4 (yellow line), 0.8 (green line), and 1 (red line). The curves are obtained numerically by solving the self-consistency equations in Eq. (24) and (25) and using the expression for the heat capacity in Eq. (26). The dash-dotted line is the result of Bethe ansatz from [20].
\[ s = \int \frac{d^2k}{V} \frac{1}{\exp [(2 (J + J') s + 2Ju_x \varepsilon_1 (k) + 2Ju_y \varepsilon_2 (k)) / (k_B T)] + 1} - \frac{1}{2}, \quad (24) \]

\[ u_\alpha = \frac{1}{2} - \int \frac{d^2k}{V} \frac{\varepsilon_\alpha (k)}{\exp [(2 (J + J') s + 2Ju_x \varepsilon_1 (k) + 2Ju_y \varepsilon_2 (k)) / (k_B T)] + 1}, \quad (25) \]

The above expression with the solutions of Eqs. (24) and (25) is plotted as a function of the anisotropy parameter \( J'/J \) at \( T = 0 \) in Fig. 5. In the 1D limit \( (J'/J = 0) \) it reproduces closely the exact value obtained using the available Bethe ansatz approach as in Section 5. In the 2D limit \( (J'/J = 1) \) the next-neighbour correlation function \( \langle S_0 \cdot S_x \rangle = \langle S_0 \cdot S_y \rangle = -0.2431 \ldots \) is reduced from its classical value \(-1/4\) due to quantum fluctuations. This is consistent with the Quantum Monte Carlo result \( \langle S_0 \cdot S_x \rangle = -0.133 \ldots \) obtained in [34]. Also for the square lattice there is no quantum phase transition at any intermediate anisotropy.

### 7 Fitting experimental data

Now, we perform a quantitative test of the theory developed in Sections 3 and 4 in the quasi-1D regime by comparing it with the experimental data for heat capacity of the anisotropic quantum antiferromagnets \( \text{Cs}_2\text{CuBr}_4 \) and \( \text{Cs}_2\text{CuCl}_4 \) on the triangular lattice measured in [3] and [35] respectively. The experimental data from these two works is plotted in Fig. 6. Blue stars are \( \text{Cs}_2\text{CuBr}_4 \) and green stars are \( \text{Cs}_2\text{CuCl}_4 \).

Both materials become 3D magnets below a critical temperature that corresponds to a weak Heisenberg coupling between the planes, \( T^{3D}_c = 2.00 \text{ K} \) for \( \text{Cs}_2\text{CuBr}_4 \) and \( T_c = 0.70\text{K} \) for \( \text{Cs}_2\text{CuCl}_4 \). Thus, we exclude the data for \( T < T^{3D}_c \) from the comparison. We also exclude the data for \( T > 0.35J/k_B \) since the spin Hartree-Fock approach has a larger quantitative discrepancy in this intermediate region of temperatures, see comparison with the exact result of Bethe ansatz in Fig. 6. We also exclude the data at very high temperatures from the fitting procedure since the lattice contribution to the heat capacity in experiments becomes appreciable there making direct comparison of the theory developed in this work for purely magnetic systems inadequate.

We use the result in Eqs. (11), (12), and (19) to fit the experimental data for both materials in intermediate range of temperatures, \( T^{3D}_c < T < 0.35J/k_B \), using \( J \) and the degree of anisotropy \( J'/J \) as the only two free fitting parameters, see red and purple lines in Fig. 6. The best fits give \( J = 1.15 \pm 0.01 \text{meV} \) and \( J'/J = 0.42 \pm 0.03 \) for \( \text{Cs}_2\text{CuBr}_4 \) and \( J = 0.37 \pm 0.01 \text{meV} \) and \( J'/J = 0.37 \pm 0.03 \) for \( \text{Cs}_2\text{CuCl}_4 \) [36]. This procedure was implemented via a minimisation routine using MATHEMATICA, giving also relatively small error bars since the fitting was done well in the nonlinear regime of intermediate temperatures for these materials. The obtained values of microscopic parameters match well the already known results of neutron
Figure 6. Comparison of the temperature dependence of the heat capacity measured experimentally at zero magnetic field for Cs$_2$CuBr$_4$ and Cs$_2$CuCl$_4$ in the quasi-1D regime with the theory based on the spin Hartree-Hartree approach developed in this paper. Blue stars are the experimental data for Cs$_2$CuBr$_4$ taken from [35]. Green stars are the experimental data for Cs$_2$CuCl$_4$ taken from [35]. Red and purple solid lines are the heat capacity evaluated for anisotropic triangular lattice using Eqs. (11), (12), and (19) for the microscopic parameters $J = 0.37 \text{meV}$, $J'/J = 0.37$ and $J = 1.15 \text{meV}$, $J'/J = 0.42$ respectively.

scattering experiments for these materials at high magnetic fields, where these microscopic parameters of the Heisenberg model are measured directly [18,2]. Thus, accuracy of the theory for heat capacity of quantum antiferromagnets in the quasi-1D regime developed in this work is confirmed.

8 Conclusions

We have applied the spin Hartree-Fock approach to the model of the anisotropic Heisenberg antiferromagnet for spin-1/2 on the square and triangular lattices. We have constructed a theory for heat capacity in the quasi-1D regime that is free of any spurious phase transition at finite temperatures, unlike the commonly used Takahashi’s modified spin-wave theory [8] and the mean-field theory based on the Schwinger-boson representation of spinwaves by Arovas and Auerbach [9]. We have successfully tested the accuracy of our newly developed approach by fitting the available data on the temperature dependence of heat capacity for a pair of anisotropic antiferromagnetic isolators [4,35] in the quasi-1D regime and obtained the microscopic exchange energies that match the values found in the neutron scattering experiments on these materials, confirming the validity of our theory.

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9 Authors contributions

All authors contributed equally to the paper.

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