



Spin Gap in β -TeVO₄: a Quantum Monte Carlo Study

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Abstract

In this paper, we proposed a two-leg spin ladder model for the description of magnetic properties of the β -TeVO₄ compound. Quantum Monte Carlo (QMC) simulation was applied to describe the temperature-dependent magnetic susceptibility data for low temperatures. The two-leg spin ladder model presents a spin gap, and we suggest that β -TeVO₄ compound presents such a spin gap also, and therefore, the model proposed here can be experimentally tested by measuring the spin gap of the compound. The susceptibility phase diagram has a rounded peak in the vicinity of $T \approx 12.2$ K and obeys Troyer's law for low temperatures and Curie's law for high temperatures. We also study the susceptibility diagram in low temperatures and found the spin gap $\Delta = 8.06$ K. The linearization of the equation for susceptibility in low temperatures allows us to obtain the spin gap value, and such a linearization was made with the data from the QMC simulation. In all the results, there is a very good agreement with the experimental data. We also show that the spin gap is null and the susceptibility is proportional to T for low temperatures when relatively high values of the ladders' coupling is considered. The theoretical results are compared with other studies as well as applied to describe the susceptibility phase diagram of consolidated spin ladder compound, C₉H₁₈N₂CuBr₄.

Keywords Magnetic frustration · Spin ladder · QMC simulation · Spin gap

1 Introduction

Low-dimensional magnetic systems present different and interesting exotic magnetic properties, and the study of the physical behavior in these systems has attracted the attention of many researchers in the recent years [1–3, 6, 9]. Complex phase diagrams, exotic magnetic behavior, and the fact that these compounds can be used as a laboratory of the quantum spin models have been some of the reasons for an increase in the studies of these systems. Among

these systems are the spin ladders: α -TeVO₄, β -TeVO₄, (C₅H₁₂N)₂CuBr₄ (bis(piperidinium) tetrabromocuprate(II), also known as BPCB), C₉H₁₈N₂CuBr₄, (VO)₂P₂O₇, SrCu₂O₃, CaCu₂O₃, whose phase diagrams depend on the composition of each element and on exchange couplings [1–9, 11]. One of the open questions about such systems is whether or not a two-leg spin ladder compound will exhibit superconducting properties and under which physical conditions [12, 13, 29].

A spin ladder, with two or more legs, in this work presents as a prototype of a quasi-unidimensional system that exhibits properties that are between one and bi-dimensional systems [8, 9, 14]. In high magnetic fields, the temperature-dependent magnetic susceptibility is written in terms of a universal scaling function [14]. Such antiferromagnetic ladders with a spin gap Δ are generally characterized by a gap in their spin excitation spectra [15], and this spin gap, Δ , is the difference between the energies of the lowest excited state and the ground state [15–17].

In general, spin ladders with an even number of legs (simplest spin ladder model) have a short-range magnetic order and a finite energy gap and are formed by two chains of spins which interact strongly via an exchange coupling $J_{\perp}$, and the spins interact with their neighboring chain

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through an exchange coupling $J_{\parallel}$ [8, 14]. The model most used to describe the magnetic properties of the β -TeVO₄ compound has been the zigzag Heisenberg chain [18, 19]. Until then, there is not any theoretical or experimental studies that consider β -TeVO₄ like a spin ladder [1, 18–21]. Therefore, description of a possible spin gap Δ for this material does not exist [1, 18, 22]. In this paper, we propose to describe theoretically the β -TeVO₄ compound with a two-leg spin ladder model, thereby obtaining a gap (Δ) that can be subsequently verified experimentally.

In particular, the zigzag Heisenberg model has been applied to describe the phase diagrams of β -TeVO₄ [1, 18, 19] and, in particular, the susceptibility and magnetization diagrams, but these models take into account the exchange interactions between the first and second nearest neighbors so as to be able to fit the experimental data to the modelling used.

In literature, there is no description of the spin gap for this compound, and here, we propose a mechanism to test experimentally the two-leg spin ladder model used to estimate the gap value. The zigzag spin 1/2 chain, with nearest and next-nearest neighbors interactions J_1 – J_2 , can be seen like a two-leg spin ladder with the conventional intra and inter chains (see [10] 10, [31] 31, [25], [26] and [28] for more details). This way, we use the spin ladder model with two legs. In this work, we map the J_1 – J_2 and use the two-leg spin ladder model with only intra ($J_{\parallel}$) e inter chain ($J_{\perp}$) interactions. This type of mapping was done to describe the magnetic properties of the CaV₂O₅ compound, in some references considered as a zigzag chain [32] and in another considered as two-leg spin ladder [33].

Initially, we discuss the results obtained for various $J_{\perp}/J_{\parallel}$ values and then we compare the results obtained for the C₉H₁₈N₂CuBr₄ compound (susceptibility diagram). In order to theoretically obtain the spin gap in β -TeVO₄, we propose using an antiferromagnetic spin 1/2 Heisenberg two-leg spin ladder model in the presence of a magnetic field for ladders with $J_{\parallel}$ and $J_{\perp}$. (They are the dominant interactions. See Savina et al. [18, 19].) The article organization is as follows: in Section 2, the simulation method and results are presented. In Section 3, some concluding observations are presented.

2 Methods and Results

For the simulation of physical properties of β -TeVO₄, we use the Heisenberg model for a spin ladder with two legs coupled for $H > 0$ [16]:

$$\mathcal{H} = J_{\parallel} \sum_{i,j=1,2} \mathbf{S}_{i,j} \cdot \mathbf{S}_{i+1,j} + J_{\perp} \sum_i \mathbf{S}_{i,1} \cdot \mathbf{S}_{i,2} - g\mu_B H \sum_i (S_{i,1}^z + S_{i,2}^z), \quad (1)$$

The indexes 1 and 2 refer to the contents of each chain, S_i^{ν} ($\nu = x, y, z$) represents the Pauli matrices at the sites i , j is the index associated with the step, H is the externally applied magnetic field, g is the Landé factor, and μ_B is the Bohr magneton. The first term of Hamiltonian (1) is linked to coupling between spins in the same chain, the second term is related to the coupling of the spins in different legs, and last term is concerning to the external magnetic field applied to the system. This model can be extended to similar systems such as the three-leg and four-leg spin ladders, for example. There is the possibility of more two interactions, three-dimensional coupling J_{3D} , and interaction from Dzyaloshinskii–Moriya type [14, 18, 23]. However, they are much smaller than $J_{\parallel}$ [24] and so will not be taken into consideration in our analysis.

We investigated the susceptibility behavior of the weak magnetic field and $H = 0$ T in order to obtain spin gap. To determine the spin gap, the magnetic susceptibility experiment was simulated using the Quantum Monte Carlo (QMC). The QMC simulations were made through the Stochastic Series Expansion (SSE) representation of the associated path integral for a mixed (ferro-antiferromagnetic) ladder system with two legs [27]. The values for $J_{\perp}$ and $J_{\parallel}$ were obtained from reference [19] and reflect well a major feature of spin ladders. The QMC simulations were performed for two-leg ladders with a size of $L = 256$ (256×2), where L is the number of spins in each leg, under periodic boundary conditions, with up to 1,000,000 steps for balance and 2,000,000 steps to the measurements. The magnetization M and the susceptibility χ were obtained using [27]

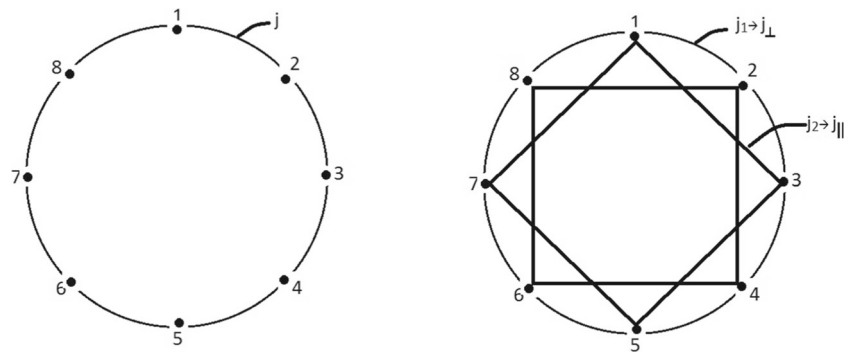
$$M = \frac{\langle \sum_i S_i^z \rangle}{N}, \quad (2)$$

$$\chi = \beta \frac{\langle (\sum_i S_i^z)^2 \rangle}{N}, \quad (3)$$

where k_B is Boltzmann's constant, $\beta \equiv 1/k_B T$, and N is spin numbers of the system.

Figure 1 displays the configurations from path Exchange Js in isotropic (J) and anisotropic (J_1 , J_2) form. The mapping done by the authors from reference [10] consists of decimating (real space renormalization group decimation process) the spin chains dividing in blocks and reducing the degrees of freedom of the system. Observe that in dividing the chain in zigzag in two blocks, there exist interactions between the spins of the same block and between spins of different blocks. When mapping the chain using a model of spin ladder, we considered the interactions between spins of the same block as being the interactions of spins belonging to the same chain and the interactions between spins of different blocks as being the interactions between spins of different chains, therefore the choice of $J_{\parallel}$ as being J_2 and $J_{\perp}$ as being J_1 , as shown the Fig. 1.

Fig. 1 Left side shows isotropic chain (isotropic J) of path exchange and right side anisotropic exchange J_1 and J_2



The graphs of Figs. 2 and 3 show a comparison between the results obtained from the simulation of two-leg spin ladders, using the model proposed by (1) and results already known in the literature, in a comparative analysis with the susceptibility data of the $\text{C}_9\text{H}_{18}\text{N}_2\text{CuBr}_4$ compound. One of the characteristics of the susceptibility behavior of two-leg spin ladders is a flattening of their peak with increasing ratio $J_\perp/J_\parallel$ as predicted by Tonel in Ref. [29]. In Fig. 2, we can see that there is excellent agreement between the proposed theoretical model and the experimental data in all temperature range. Integrable spin ladder models generally disagree with experimental data at high temperatures. In the simulations, for $\text{C}_9\text{H}_{18}\text{N}_2\text{CuBr}_4$, the values of $J_\perp$ and $J_\parallel$ used were extracted from reference [5].

The analysis from temperature-dependent magnetic susceptibility (Fig. 4) shows the typical magnetic characteristics of spin ladders: There is a rounded peak around the magnetic transition temperature (12.2 K) [8, 14]. In low temperatures, the susceptibility decays exponentially in accordance with Troyer's law (Fig. 5). The spin ladders were considered in isolation, i.e., $J_{2D} \ll J_\perp, J_\parallel$, where J_{2D} is the exchange coupling between distinct ladders [8, 9, 14, 18,

19]. When $J_\perp \approx J_\parallel \approx J_{2D}$, this system presents a spin 1/2 Heisenberg model in a squared lattice behavior [8, 9].

There are many ways to obtain the exchange values. In Savina et al. [19], the values of the exchange couplings were obtained by adjusting the susceptibility curve, $\chi(J_1/J_2)$, and by the magnitude and the temperature position of the maximum of the magnetic susceptibility in function from ratio $J_\parallel/J_\perp$, $T_{\max}(J_\parallel/J_\perp)$. In this work, we used the values obtained in the cited paper: $J_\perp = 29.5$ K and $J_\parallel = -38.3$ K [19]. This is a typical empirical adjustment, often present in theoretical and/or experimental studies of spin ladders [8, 9]. The best fit from experimental data of the susceptibility is obtained when we assume an empirical relation $T\chi_{\max} \propto |J_\parallel|$ [8, 18, 19]. The value of $T\chi_{\max}$ would be related to the spin gap Δ/k_B of the system, which can easily be obtained from the fit of the susceptibility curve in low temperatures, allowing us to calculate the exchange values [30]. As can be seen in Fig. 4, the agreement between

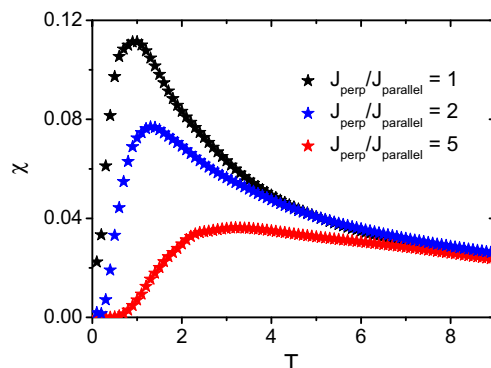


Fig. 2 Temperature dependence of the theoretical magnetic susceptibility (arbitrary units) for $H = 0T$. We use three $J_\perp/J_\parallel$ ($J_{\text{perp}} = J_\perp$ and $J_{\text{parallel}} = J_\parallel$) ratio values. We can observe a flattening in the peaks and an increase in the transition temperature in the function of the ratio $J_\perp/J_\parallel$. This result is in excellent agreement with that obtained from Reference [29]

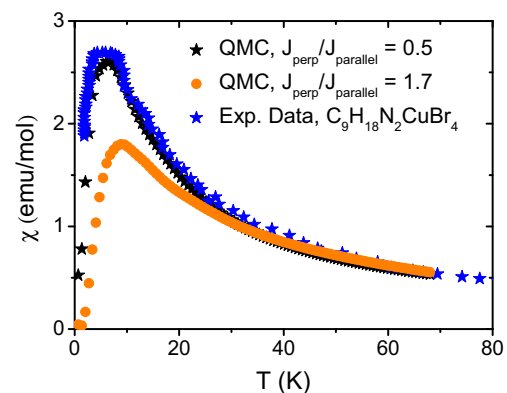


Fig. 3 Susceptibility phase diagram (and experimental data for $\text{C}_9\text{H}_{18}\text{N}_2\text{CuBr}_4$) with two $J_\perp/J_\parallel$ ($J_{\text{perp}} = J_\perp$ and $J_{\text{parallel}} = J_\parallel$) ratio values. The experimental data were obtained from Awwadi et al. [5] and the QMC data were obtained via (3). The concordance between simulation and experiment is excellent and the behavior of the susceptibility in the limits of high and low temperatures obeys (5) and (4), respectively. In high temperature, all models (also experimental data) behave as described by (5). The exchange interactions are (antiferromagnetic, positive sign) $J_\perp = 7.95$ and $J_\parallel = 4.1$, $J_\perp/J_\parallel \approx 0.5$

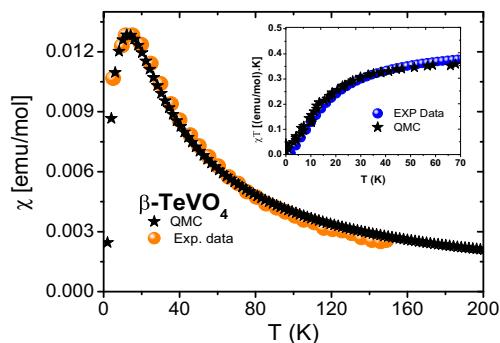


Fig. 4 Magnetic susceptibility in magnetic field $H = 0.1$ T. The experimental data were obtained from Pregelj et al. [1], and the QMC data were obtained via (3). The concordance between simulation and experiment is excellent, and the behavior of the susceptibility in the limits of high and low temperatures obeys the (5) and (4) respectively. The inset shows $\chi(T)T$ for single crystal of β -TeVO₄ fit experimental data using the QMC simulation. The estimate of $T\chi_{\max}$ allows us to obtain the values of the exchange parameters [19]

the results obtained from fits and the experimental data is excellent in all temperature range.

There are some proposals for the expressions of the calculation (susceptibility and gap spin), as used by Tonel et al. [29], that also investigated the behavior of the susceptibility of BPCB through a model with an exactly soluble spin ladder via the Bethe ansatz, but the agreement with the experimental results was only good at low temperatures. For this, the authors considered $\Delta = J_{\perp} - J_{\parallel}$ for BPCB. Here, we have used the analysis from temperature dependence susceptibility using Troyer's law [29], considering that the susceptibility decays exponentially,

$$\chi = C \cdot \frac{e^{-\Delta/T}}{\sqrt{T}}, \quad (4)$$

where C is a constant, Δ is the spin gap, and T is the temperature. The exponential decay at low temperatures

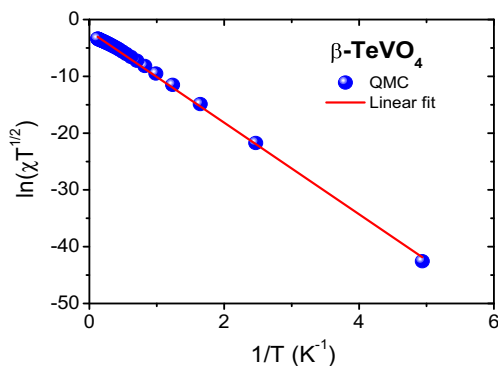


Fig. 5 QMC simulations from magnetic susceptibility in low temperature range for the determination of spin gap (Δ). By using (4), the linearization provided $\Delta = 8.06$ K. The experimental data are from Pregelj et al. [1]

indicates the presence of a spin gap [29]. Chains with an odd number of legs do not exhibit this kind of behavior at low temperatures [14]. The linearization of (4) allows us to obtain the value of the spin gap, and the linearization was made with the data from the QMC simulation and can be seen in Fig. 5. The gap obtained via the linear fit is $\Delta = 8.06$ K, a small value when compared to another compounds [9].

Deriving the (4), we get

$$-\frac{\partial}{\partial \beta} \ln \chi = \Delta - \frac{1}{2}T,$$

which physically it means that for systems where the rate of change of $\ln \chi$ goes to zero (one-dimensional magnetic systems, for example) the gap disappears when $T \rightarrow 0$, even though it remains finite on spin ladders. The agreement between experiment and theory persists in this paper even at high temperatures ($T > T_N$), as can be seen in the inset of Fig. 4. At high temperatures, the susceptibility follows Curie's law [14]:

$$\chi \propto \frac{1}{T} \quad (5)$$

Finally, it is important to note that results show excellent agreement between the experimental data and the QMC in all temperature range analyzed.

3 Concluding Remarks

In this paper, we presented the results of QMC simulations of the two-leg spin ladder model with applications in the description of magnetic properties of C₉H₁₈N₂CuBr₄ and β -TeVO₄ compounds and the theoretical prediction of the spin gap from β -TeVO₄ compound, being the experimental verification of this spin gap a test criterion of the model proposed here. In this work, we map a zigzag chain in a spin ladder model as described in reference [10]. We study the diagram from magnetic susceptibility in low magnetic field, with null magnetic field and for some values of interactions $J_{\perp}$ and $J_{\parallel}$, the susceptibility phase diagram at low temperatures, and the $\chi(T)T$ product. Based on the susceptibility diagram at low temperatures, we found, using a spin ladder model, a spin gap value of $\Delta = 8.06$ K. The two-leg Heisenberg (with the parameters described in the text) predicts the existence of a small gap. We believe that further experimental studies are necessary to test the hypothesis that the β -TeVO₄ compound (its magnetic structure) can be mapped from a zigzag Heisenberg chain to a spin ladder with two legs.

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